

CASE 1-22224/US/A

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

Group Art Unit: 1714

FRANÇOIS GUGUMUS

Examiner:

APPLICATION NO: 09/899,438

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FILED: JULY 5, 2001

FOR: STABILIZER MIXTURES

TC 1700

Assistant Commissioner for Patents Washington, D.C. 20231

CLAIM OF PRIORITY UNDER 35 USC 119

Sir:

Applicant in the above-entitled application by his attorney hereby claims priority under the International Convention of European application No. 00810621.3, filed on July 14, 2000, This application is acknowledged in the Declaration of the instant case.

A certified copy of said European application is submitted herewith.

Respectfully submitted.

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DRC/am/ Encl. (1)

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Eur päisches **Patentamt**

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Bescheinigung

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Attestation

Die angehefteten Unterlagen stimmen mit der ursprünglich eingereichten Fassung der auf dem nächsten Blatt bezeichneten europäischen Patentanmeldung überein.

The attached documents are exact copies of the European patent application conformes à la version described on the following initialement déposée de page, as originally filed.

Les documents fixés à cette attestation sont initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr.

Patent application No. Demande de brevet n°

00810621.3

Der Präsident des Europäischen Patentamts; Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets p.o.

I.L.C. HATTEN-HECKMAN

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Blatt 2 der Besch inigung Sheet 2 of the certificate Page 2 de l'attestation

Anmeldung Nr.: Application no.: Demande n*:

00810621.3

Anmeldetag: Date of filing: Date de dépôt:

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Anmelder: Applicant(s): Demandeur(s):

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Bezeichnung der Erfindung: Title of the invention: Titre de l'invention: Stabilizer mixtures

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Stabilizer mixtures

The present invention relates to a stabilizer mixture containing two different sterically hindered amine compounds and at least one Mg- and/or Zn- compound, the use of this mixture for stabilizing an organic material, in particular a polyolefin, against degradation induced by light, heat or oxidation and the organic material thus stabilized.

The stabilization of polyolefins is described in numerous publications, for example in US-A-4,929,652, US-A-5,025,051, US-A-5,037,870, EP-A-276,923, EP-A-290,388, EP-A-429,731, EP-A-468,923, EP-A-661,341, EP-A-690,094, DE-A-19,545,896 (Derwent 96-278,994/29; Chemical Abstracts 125:116779q), WO-A-95/25,767, GB-A-2,293,827, Chemical Abstracts 106:197407z and GB-A-2,332,678.

In more detail, the present invention relates to a stabilizer mixture containing

- (I) two different sterically hindered amine compounds, and
- (II) at least one compound selected from the group consisting of an organic salt of Zn, an inorganic salt of Zn, Zn oxide, Zn hydroxide, an organic salt of Mg, an inorganic salt of Mg, Mg oxide and Mg hydroxide.

The two different sterically hindered amine compounds of component (I) are preferably selected from the group consisting of the following classes

 $(\alpha-1)$ a compound of the formula (A-1)

$$\begin{bmatrix} H_3C & CH_3 & & & \\ E_1 & N & CH_3 & & & \\ H_3C & CH_3 & & & & \\ \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & & & \\ & & & \\ & & & \\ \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & & & \\ & & & \\ \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & & & \\ & & & \\ \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & & & \\ & & & \\ \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & & & \\ & & & \\ \end{bmatrix}$$

in which

 E_1 is hydrogen, C_1 - C_8 alkyl, O° , -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_8 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl,

m₁ is 1, 2 or 4,

if m₁ is 1, E₂ is C₁-C₂₅alkyl,

if m₁ is 2, E₂ is C₁-C₁₄alkylene or a group of the formula (a-I)

$$- \begin{matrix} \downarrow \\ C \\ E_3 \end{matrix} \qquad \begin{matrix} E_5 \\ E_8 \end{matrix} \qquad (a-l)$$

wherein E_3 is C_1 - C_{10} alkyl or C_2 - C_{10} alkenyl, E_4 is C_1 - C_{10} alkylene, and E_5 and E_6 independently of one another are C_1 - C_4 alkyl, cyclohexyl or methylcyclohexyl, and

if m₁ is 4, E₂ is C₄-C₁₀alkanetetrayl;

 $(\alpha-2)$ a compound of the formula (A-2)

in which

two of the radicals E_7 are -COO-(C_1 - C_{20} alkyl), and two of the radicals E_7 are a group of the formula (a-II)

$$-- COO - H_3C CH_3$$

with E₈ having one of the meanings of E₁;

 $(\alpha-3)$ a compound of the formula (A-3)

in which

 E_9 and E_{10} together form C_2 - C_{14} alkylene, E_{11} is hydrogen or a group - Z_1 -COO- Z_2 , Z_1 is C_2 - C_{14} alkylene, and Z_2 is C_1 - C_{24} alkyl, and

E₁₂ has one of the meanings of E₁;

 $(\alpha-4)$ a compound of the formula (A-4)

wherein

the radicals E_{13} independently of one another have one of the meanings of E_1 , the radicals E_{14} independently of one another are hydrogen or C_{1} - C_{12} alkyl, and E_{15} is C_1 - C_{10} alkylene or C_3 - C_{10} alkylidene;

 $(\alpha-5)$ a compound of the formula (A-5)

wherein

the radicals E₁₆ independently of one another have one of the meanings of E₁;

 $(\alpha-6)$ a compound of the formula (A-6)

in which

E₁₇ is C₁-C₂₄alkyl, and

E₁₈ has one of the meanings of E₁;

 $(\alpha-7)$ a compound of the formula (A-7)

$$E_{19} \longrightarrow E_{21}$$

$$E_{20} \longrightarrow E_{20}$$
(A-7)

in which

 E_{19} , E_{20} and E_{21} independently of one another are a group of the formula (a-III)

$$--CH_{2}-CH-CH_{2}-NH-CH_{3}-N-E_{22}$$

$$OH$$

$$H_{3}C$$

$$CH_{3}$$

$$(a-III)$$

wherein E22 has one of the meanings of E1;

$(\alpha-8)$ a compound of the formula (A-8)

wherein

the radicals E_{23} independently of one another have one of the meanings of E_1 , and E_{24} is hydrogen, C_1 - C_{12} alkyl or C_1 - C_{12} alkoxy;

(α-9) a compound of the formula (A-9)

$$\begin{bmatrix} H_3C & CH_3 & O \\ E_{\overline{25}} & N & N & E_{26} \\ H_3C & CH_3 & m_2 \end{bmatrix}$$
 (A-9)

wherein

m₂ is 1, 2 or 3,

E25 has one of the meanings of E1, and

when
$$m_2$$
 is 1, E_{26} is a group $--CH_2CH_2-NH-\langle \rangle$,

when m_2 is 2, E_{26} is C_2 - C_{22} alkylene, and when m_2 is 3, E_{26} is a group of the formula (a-IV)

wherein the radicals E_{27} independently of one another are C_2 - C_{12} alkylene, and the radicals E_{28} independently of one another are C_1 - C_{12} alkyl or C_5 - C_{12} cycloalkyl;

(α-10) a compound of the formula (A-10)

wherein

the radicals E_{29} independently of one another have one of the meanings of E_1 , and E_{30} is C_2 - C_{22} alkylene, C_5 - C_7 cycloalkylene, C_1 - C_4 alkylenedi(C_5 - C_7 cycloalkylene), phenylenedi(C_1 - C_4 alkylene);

(β-1) a compound of the formula (B-1)

in which

 R_1 , R_3 , R_4 and R_5 independently of one another are hydrogen, C_1 - C_{12} alkyl, C_5 - C_{12} cycloalkyl, C_1 - C_4 -alkyl-substituted C_5 - C_{12} cycloalkyl, phenyl, phenyl which is substituted by -OH and/or C_1 - C_{10} alkyl; C_7 - C_9 phenylalkyl, C_7 - C_9 phenylalkyl which is substituted on the phenyl radical by -OH and/or C_1 - C_{10} alkyl; or a group of the formula (b-l)

$$H_3C$$
 CH_3 H_3C CH_3 (b-i)

 R_2 is C_2 - C_{18} alkylene, C_5 - C_7 cycloalkylene or C_1 - C_4 alkylenedi(B_5 - C_7 cycloalkylene), or

the radicals R_1 , R_2 and R_3 , together with the nitrogen atoms to which they are bonded, perform a 5- to 10-membered heterocyclic ring, or

R₄ and R₅, together with the nitrogen atom to which they are bonded, form a 5- to 10-membered heterocyclic ring,

 R_6 is hydrogen, C_1 - C_8 alkyl, O^{\cdot} , -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl, and

b₁ is a number from 2 to 50,

with the proviso that at least one of the radicals R_1 , R_3 , R_4 and R_5 is a group of the formula (b-1);

 $(\beta-2)$ a compound of the formula (B-2)

wherein

R₇ and R₁₁ independently of one another are hydrogen or C₁-C₁₂alkyl,

R₈, R₉ and R₁₀ independently of one another are C₂-C₁₀alkylene, and

 X_1 , X_2 , X_3 , X_4 , X_5 , X_6 , X_7 and X_8 independently of one another are a group of the formula (b-II),

in which R_{12} is hydrogen, C_1 - C_{12} alkyl, C_5 - C_{12} cycloalkyl, C_1 - C_4 alkyl-substituted C_5 - C_{12} cycloalkyl, phenyl, -OH- and/or C_1 - C_{10} alkyl-substituted phenyl, C_7 - C_9 phenylalkyl which is substituted on the phenyl radical by -OH and/or C_1 - C_{10} alkyl; or a group of the formula (b-l) as defined above, and R_{13} has one of the meanings of R_6 ;

(β-3) a compound of the formula (B-3)

(B-3)

in which

 R_{14} is C_1 - C_{10} alkyl, C_5 - C_{12} cycloalkyl, C_1 - C_4 alkyl-substituted C_5 - C_{12} cycloalkyl, phenyl or C_1 - C_{10} alkyl-substituted phenyl,

R₁₅ is C₃-C₁₀alkylene,

R₁₆ has one of the meanings of R₆, and

b₂ is a number from 2 to 50;

(β-4) a compound of the formula (B-4)

in which

 R_{17} and R_{21} independently of one another are a direct bond or a -N(X₉)-CO-X₁₀-CO-N(X₁₁)-group, where X₉ and X₁₁ independently of one another are hydrogen, C₁-C₈alkyl,

C₅-C₁₂cycloalkyl, phenyl, C₇-C₉phenylalkyl or a group of the formula (b-l),

X₁₀ is a direct bond or C₁-C₄alkylene,

R₁₈ has one of the meanings of R₆,

R₁₉, R₂₀, R₂₃ and R₂₄ independently of one another are hydrogen, C₁-C₃₀alkyl,

C₅-C₁₂cycloalkyl or phenyl,

R₂₂ is hydrogen, C₁-C₃₀alkyl, C₅-C₁₂cycloalkyl, phenyl, C₇-C₉phenylalkyl or a group of the formula (b-I), and

b₃ is a number from 1 to 50;

 $(\beta-5)$ a compound of the formula $(\beta-5)$

in which

 R_{25} , R_{26} , R_{27} , R_{28} and R_{29} independently of one another are a direct bond or C_1 - C_{10} alkylene,

 R_{30} has one of the meanings of R_6 , and b_4 is a number from 1 to 50;

 $(\beta-6)$ a product (B-6) obtainable by reacting a product, obtained by reaction of a polyamine of the formula (B-6-1) with cyanuric chloride, with a compound of the formula (B-6-2)

$$H_2N - (CH_2)_{\overline{b_5}} NH - (CH_2)_{\overline{b_5}} NH - (CH_2)_{\overline{b_5}} NH_2$$
 (B-6-1)

$$H_3C$$
 H_3C
 H_3C

in which

b'₅, b"₅ and b"'₅ independently of one another are a number from 2 to 12, R_{31} is hydrogen, C_1 - C_{12} alkyl, C_5 - C_{12} cycloalkyl, phenyl or C_7 - C_9 phenylalkyl, and R_{32} has one of the meanings of R_6 ;

 $(\beta-7)$ a compound of the formula (B-7)

wherein A_1 is hydrogen or C_1 - C_4 alkyl, A_2 is a direct bond or C_1 - C_{10} alkylene, and n_1 is a number from 2 to 50;

 $(\beta-8)$ at least one compound of the formulae (B-8-a) and (B-8-b)

$$\begin{array}{c|c}
\hline
CH & CH_2 & O \\
\hline
CH_2 & O & O \\
\hline
CH_2 & O & O \\
\hline
CH_3 & CH_3 & O \\
\hline
H_3C & CH_3 & O \\
\hline
CH_3 & CH_3 & O \\
\hline
CH_3 & O & O \\
\hline
CH_4 & O & O \\
\hline
CH_5 & O & O \\
\hline
CH_6 & O & O \\
\hline
CH_7 & O & O \\
\hline
C$$

wherein n₂ and n₂* are a number from 2 to 50;

 $(\beta-9)$ a compound of the formula (B-9)

(B-9)

wherein A_3 and A_4 independently of one another are hydrogen or C_1 - C_8 alkyl, or A_3 and A_4 together form a C_2 - C_{14} alkylene group, and

the variables n₃ independently of one another are a number from 1 to 50; and

$(\beta-10)$ a compound of the formula (B-10)

wherein n₄ is a number from 2 to 50,

A₅ is hydrogen or C₁-C₄alkyl,

the radicals A_6 and A_7 independently of one another are C_1 - C_4 alkyl or a group of the formula (b-l),

with the proviso that at least 50 % of the radicals A₇ are a group of the formula (b-l).

Examples of component (I) are:

- A compound selected from the class α -1 and a compound selected from the class α -3, α -5, α -6, α -7, α -9, α -10, β -1, β -2, β -3, β -4, β -5, β -6, β -7, β -8 or β -9.
- A compound selected from the class α-3 and a compound selected from the class α-5,
 α-6, α-7, α-9, α-10, β-1, β-2, β-3, β-4, β-5, β-6, β-7, β-8 or β-9.

- A compound selected from the class α-5 and a compound selected from the class α-6,
 α-7, α-9, α-10, β-1, β-2, β-3, β-4, β-5, β-6, β-7, β-8 or β-9.
- A compound selected from the class α-6 and a compound selected from the class α-9,
 α-10, β-1, β-2, β-3, β-4, β-5, β-6, β-7, β-8 or β-9.
- A compound selected from the class α-7 and a compound selected from the class α-9,
 α-10, β-1, β-2, β-3, β-4, β-5, β-6, β-7, β-8 or β-9.
- A compound selected from the class α-10 and a compound selected from the class β-1,
 β-2, β-3, β-4, β-5, β-6, β-7, β-8 or β-9.
- A compound selected from the class β-1 and a compound selected from the class β-2,
 β-3, β-4, β-5, β-6, β-7, β-8, β-9 or β-10.
- A compound selected from the class β-2 and a compound selected from the class β-3,
 β-4, β-5, β-6, β-7, β-8, β-9 or β-10.
- A compound selected from the class β-3 and a compound selected from the class β-4,
 β-5, β-6, β-7, β-8, β-9 or β-10.
- A compound selected from the class β-4 and a compound selected from the class β-5,
 β-6, β-7, β-8, β-9 or β-10.
- A compound selected from the class β-5 and a compound selected from the class β-6,
 β-7, β-8, β-9 or β-10.
- A compound selected from the class β-6 and a compound selected from the class β-7,
 β-8, β-9 or β-10.
- A compound selected from the class β-7 and a compound selected from the class β-8,
 β-9 or β-10.
- A compound selected from the class β-8 and a compound selected from the class β-9 or β-10.
- A compound selected from the class β-9 and a compound selected from the class β-10.

According to a preferred embodiment,

the two different sterically hindered amine compounds of component (I) are selected from the group consisting of the classes $(\alpha-1)$, $(\alpha-2)$, $(\alpha-3)$, $(\alpha-4)$, $(\alpha-5)$, $(\alpha-6)$, $(\alpha-7)$, $(\alpha-8)$, $(\alpha-9)$ and $(\alpha-10)$.

According to a further preferred embodiment,

the two different sterically hindered amine compounds of component (I) are selected from the group consisting of the classes (β -1), (β -2), (β -3), (β -4), (β -5), (β -6), (β -7), (β -8), (β -9) and (β -10).

According to another preferred embodiment,

one of the two different sterically hindered amine compounds of component (I) is selected from the group consisting of the classes (α -1), (α -2), (α -3), (α -4), (α -5), (α -6), (α -7), (α -8), (α -9) and (α -10), and

the other of the two different sterically hindered amine compounds of component (I) is selected from the group consisting of the classes (β -1), (β -2), (β -3), (β -4), (β -5), (β -6), (β -7), (β -8), (β -9) and (β -10).

One of the two different sterically hindered amine compounds of component (I) is preferably selected from the class (β -1).

According to a particularly preferred embodiment, one of the two different sterically hindered amine compounds of component (I) is selected from the class (β -1), and the other of the two different sterically hindered amine compounds of component (I) is selected from the class (α -1) or (β -7).

According to another particularly preferred embodiment, one of the two different sterically hindered amine compounds of component (I) is selected from the class (β -7), and the other of the two different sterically hindered amine compounds of component (I) is selected from the class (β -2).

The two different sterically hindered amine compounds of component (I) are preferably selected from different classes.

Examples of alkyl having up to 30 carbon atoms are methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethyl-

butyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, eicosyl, docosyl and triacontyl. One of the preferred definitions of E₁, E₈, E₁₂, E₁₃, E₁₆, E₁₈, E₂₂, E₂₃, E₂₅, E₂₉, R₆, R₁₃, R₁₆, R₁₉, R₃₀ and R₃₂ is C₁-C₄alkyl, especially methyl. R₃₁ is preferably butyl.

Examples of alkoxy having up to 18 carbon atoms are methoxy, ethoxy, propoxy, isopropoxy, butoxy, isobutoxy, pentoxy, isopentoxy, hexoxy, heptoxy, octoxy, decyloxy, dodecyloxy, tetradecyloxy, hexadecyloxy and octadecyloxy. One of the preferred meanings of E_1 is octoxy. E_{24} is preferably C_1 - C_4 alkoxy and one of the preferred meanings of R_6 is propoxy.

Examples of C₅-C₁₂cycloalkyl are cyclopentyl, cyclohexyl, cycloheptyl, cycloactyl and cyclododecyl. C₅-C₈Cycloalkyl, especially cyclohexyl, is preferred.

C₁-C₄Alkyl-substituted C₅-C₁₂cycloalkyl is for example methylcyclohexyl or dimethylcyclohexyl.

Examples of C₅-C₁₂cycloalkoxy are cyclopentoxy, cyclohexoxy, cycloheptoxy, cyclooctoxy, cyclodecyloxy and cyclodecyloxy. C₅-C₈Cycloalkoxy, in particular cyclopentoxy and cyclohexoxy, is preferred.

-OH- and/or C₁-C₁₀alkyl-substituted phenyl is for example methylphenyl, dimethylphenyl, trimethylphenyl, tert-butylphenyl or 3,5-di-tert-butyl-4-hydroxyphenyl.

Examples of C₇-C₉phenylalkyl are benzyl and phenylethyl.

C₇-C₉Phenylalkyl which is substituted on the phenyl radical by -OH and/or by alkyl having up to 10 carbon atoms is for example methylbenzyl, dimethylbenzyl, trimethylbenzyl, tert-butylbenzyl or 3,5-di-tert-butyl-4-hydroxybenzyl.

Examples of alkenyl having up to 10 carbon atoms are allyl, 2-methallyl, butenyl, pentenyl and hexenyl. Allyl is preferred. The carbon atom in position 1 is preferably saturated.

Examples of acyl containing not more than 8 carbon atoms are formyl, acetyl, propionyl, butyryl, pentanoyl, hexanoyl, heptanoyl, octanoyl, acryloyl, methacryloyl and benzoyl. C₁-C₈Alkanoyl, C₃-C₈alkenyl and benzoyl are preferred. Acetyl and acryloyl are especially preferred.

Examples of alkylene having up to 22 carbon atoms are methylene, ethylene, propylene, trimethylene, tetramethylene, pentamethylene, 2,2-dimethyltrimethylene, hexamethylene, trimethylene, octamethylene and decamethylene.

An example of C_3 - C_{10} alkylidene is the group $-C_{--}$.

An example of C₄-C₁₀alkanetetrayl is 1,2,3,4-butanetetrayl.

An example of C₅-C₇cycloalkylene is cyclohexylene.

An example of C₁-C₄alkylenedi(C₅-C₇cycloalkylene) is methylenedicyclohexylene.

An example of phenylenedi(C_1 - C_4 alkylene) is methylene-phenylene-methylene or ethylene-phenylene-ethylene.

Where the radicals R_1 , R_2 and R_3 , together with the nitrogen atoms to which they are attached, form a 5- to 10-membered heterocyclic ring, this ring is for example

A 6-membered heterocyclic ring is preferred.

Where the radicals R₄ and R₅, together with the nitrogen atom to which they are attached, form a 5- to 10-membered heterocyclic ring, this ring is for example 1-pyrrolidyl, piperidino, morpholino, 1-piperazinyl, 4-methyl-1-piperazinyl, 1-hexahydroazepinyl, 5,5,7-trimethyl-1-homopiperazinyl or 4,5,5,7-tetramethyl-1-homopiperazinyl. Morpholino is particularly

preferred.

One of the preferred definitions of R_{19} and R_{23} is phenyl.

R₂₆ is preferably a direct bond.

 n_1 , n_2 , n_2 * and n_4 are preferably a number from 2 to 25, in particular 2 to 20. n_3 is preferably a number from 1 to 25, in particular 1 to 20.

b₁ and b₂ are preferably a number from 2 to 25, in particular 2 to 20.

b₃ and b₄ are preferably a number from 1 to 25, in particluar 1 to 20.

b'₅ and b"'₅ are preferably 3 and b"₅ is preferably 2.

The compounds described above as components (I) and (II) are essentially known and commercially available. All of them can be prepared by known processes.

The preparation of the compounds of component (I) is disclosed, for example, in US-A-5,679,733, US-A-3,640,928, US-A-4,198,334, US-A-5,204,473, US-A-4,619,958, US-A-4,110,306, US-A-4,110,334, US-A-4,689,416, US-A-4,408,051, SU-A-768,175 (Derwent 88-138,751/20), US-A-5,049,604, US-A-4,769,457, US-A-4,356,307, US-A-4,619,956, US-A-5,182,390, GB-A-2,269,819, US-A-4,292,240, US-A-5,026,849, US-A-5,071,981, US-A-4,547,538, US-A-4,976,889, US-A-4,086,204, US-A-6,046,304, US-A-4,331,586, US-A-4,108,829, US-A-5,051,458, WO-A-94/12,544 (Derwent 94-177,274/22), DD-A-262,439 (Derwent 89-122,983/17), US-A-4,857,595, US-A-4,529,760, US-A-4,477,615, CAS 136,504-96-6, US-A-4,233,412, US-A-4,340,534, WO-A-98/51,690 and EP-A-1,803.

The product (B-6) can be prepared analogously to known processes, for example by reacting a polyamine of formula (B-6-1) with cyanuric chloride in a molar ratio

of from 1:2 to 1:4 in the presence of anhydrous lithium carbonate, sodium carbonate or potassium carbonate in an organic solvent such as 1,2-dichloroethane, toluene, xylene, benzene, dioxane or tert-amyl alcohol at a temperature of from -20°C to +10°C, preferably from -10°C to +10°C, in particular from 0°C to +10°C, for from 2 to 8 hours, followed by reaction of the resultant product with a 2,2,6,6-tetramethyl-4-piperidylamine of the formula (B-6-2). The molar ratio of the 2,2,6,6-tetramethyl-4-piperidylamine to polyamine of the formula (B-6-1) employed is for example from 4:1 to 8:1. The quantity of the 2,2,6,6-tetramethyl-4-piperidylamine can be added in one portion or in more than one portion at intervals of a few hours.

The molar ratio of polyamine of the formula (B-6-1) to cyanuric chloride to 2,2,6,6-tetramethyl-4-piperidylamine of the formula (B-6-2) is preferably from 1:3:5 to 1:3:6.

The following example indicates one way of preparing a preferred product (B-6-a).

Example: 23.6 g (0.128 mol) of cyanuric chloride, 7.43 g (0.0426 mol) of N,N'-bis[3-aminopropyl]ethylenediamine and 18 g (0.13 mol) of anhydrous potassium carbonate are reacted at 5°C for 3 hours with stirring in 250 ml of 1,2-dichloroethane. The mixture is warmed at room temperature for a further 4 hours. 27.2 g (0.128 mol) of N-(2,2,6,6-tetramethyl-4-piperidyl)butylamine are added and the resultant mixture is warmed at 60°C for 2 hours. A further 18 g (0.13 mol) of anhydrous potassium carbonate are added and the mixture is warmed at 60°C for a further 6 hours. The solvent is removed by distillation under a slight vacuum (200 mbar) and replaced by xylene. 18.2 g (0.085 mol) of N-(2,2,6,6-tetramethyl-4-piperidyl)butylamine and 5.2 g (0.13 mol) of ground sodium hydroxide are added, the mixture is heated at reflux for 2 hours and, for a further 12 hours, the water formed during the reaction is removed by azeotropic distillation. The mixture is filtered. The solution is washed with water and dried over Na₂SO₄. The solvent is evaporated and the residue is dried at 120-130°C in vacuo (0.1 mbar). The desired product is obtained as a colourless resin.

In general, the product (B-6) can, for example, be represented by a compound of

the formula (B-6- α), (B-6- β) or (B-6- γ). It can also be in the form of a mixture of these three compounds.

A preferred meaning of the formula (B-6- α) is

A preferred meaning of the formula (B-6- β) is

A preferred meaning of the formula (B-6-γ) is

In the above formulae (B-6- α) to (B-6- γ), b₅ is preferably 2 to 20, in particular 2 to 10.

The two different sterically hindered amine compounds of component (I) are preferably selected from the group consisting of the following commercial products:

DASTIB 845 (RTM), TINUVIN 770 (RTM), TINUVIN 765 (RTM), TINUVIN 144 (RTM), TINUVIN 123 (RTM), MARK LA 52 (RTM), MARK LA 57 (RTM), MARK LA 67 (RTM), HOSTAVIN N 20 (RTM), HOSTAVIN N 24 (RTM), SANDUVOR 3050 (RTM), DIACETAM 5 (RTM), SUMISORB TM 61 (RTM), UVINUL 4049 (RTM), SANDUVOR PR 31 (RTM), GOODRITE UV 3034 (RTM),

GOODRITE UV 3150 (RTM), GOODRITE UV 3159 (RTM), GOODRITE 3110 x 128 (RTM), UVINUL 4050 H (RTM), CHIMASSORB 944 (RTM), CHIMASSORB 2020 (RTM), CYASORB UV 3346 (RTM), CYASORB UV 3529 (RTM), DASTIB 1082 (RTM), CHIMASSORB 119 (RTM), UVASIL 299 (RTM), UVASIL 125 (RTM), UVASIL 2000 (RTM), UVINUL 5050 H (RTM), LICHTSCHUTZSTOFF UV 31 (RTM), LUCHEM HA B 18 (RTM), MARK LA 63 (RTM), MARK LA 68 (RTM), UVASORB HA 88 (RTM), TINUVIN 622 (RTM), HOSTAVIN N 30 (RTM) and FERRO AM 806 (RTM).

The meanings of the terminal groups which saturate the free valences in the compounds of the formulae (B-1), (B-3), (B-4), (B-5), (B-6- α), (B-6- β), (B-6- γ), (B-7), (B-8-a), (B-8-b) and (B-10) depend on the processes used for their preparation. The terminal groups can also be modified after the preparation of the compounds.

If the compounds of the formula (B-1) are prepared by reacting a compound of the formula

$$X \longrightarrow N \longrightarrow X$$

$$N \longrightarrow N$$

$$N \longrightarrow R_5$$

$$R_4$$

in which X is, for example, halogen, in particular chlorine, and R_4 and R_5 are as defined above, with a compound of the formula

in which R₁, R₂ and R₃ are as defined above, the terminal group bonded to the diamino radical is hydrogen or

and the terminal group bonded to the triazine radical is X or

If X is halogen, it is advantageous to replace this, for example, by -OH or an amino group when the reaction is complete. Examples of amino groups which may be mentioned are pyrrolidin-1-yl, morpholino, -NH₂, -N(C₁-C₈)alkyl)₂ and -NR(C₁-C₈alkyl), in which R is hydrogen or a group of the formula (b-l).

The compounds of the formula (B-1) also cover compounds of the formula

wherein R_1 , R_2 , R_3 , R_4 , R_5 and b_1 are as defined above and R_4 * has one of the meanings of R_4 and R_5 * has one of the meanings of R_5 .

One of the particularly preferred compounds of the formula (B-1) is

The preparation of this compound is described in Example 10 of US-A-6,046,304.

In the compounds of the formula (B-3), the terminal group bonded to the silicon atom can be, for example, $(R_{14})_3Si$ -O-, and the terminal group bonded to the oxygen can be, for example, $-Si(R_{14})_3$.

The compounds of the formula (B-3) can also be in the form of cyclic compounds if b₂ is a number from 3 to 10, i.e. the free valences shown in the structural formula then form a direct bond.

In the compounds of the formula (B-4), the terminal group bonded to the 2,5-dioxopyrrolidine ring is, for example, hydrogen, and the terminal group bonded to the $-C(R_{23})(R_{24})$ - radical is, for example,

In the compounds of the formula (B-5), the terminal group bonded to the carbonyl radical is, for example,

and the terminal group bonded to the oxygen radical is, for example,

In the compounds of the formulae (B-6- α), (B-6- β) and (B-6- γ), the terminal group bonded to the triazine radical is, for example, CI or a

group, and the terminal group bonded to the amino radical is, for example, hydrogen or a

group.

If the compounds of the formula (B-7) are prepared, for example, by reacting a compound of the formula

in which A₁ is hydrogen or methyl, with a dicarboxylic acid diester of the formula Y-OOC-A₂-COO-Y, in which Y is, for example, methyl, ethyl or propyl, and A₂ is as defined above, the terminal group bonded to the 2,2,6,6-tetramethyl-4-oxypiperidin-1-yl radical is hydrogen or -CO-A₂-COO-Y, and the terminal group bonded to the diacyl radical is -O-Y or

In the compounds of the formula (B-8-a), the terminal group bonded to the nitrogen can be, for example, hydrogen and the terminal group bonded to the 2-hydroxypropylene radical can be, for example, a

group.

In the compounds of the formula (B-8-b), the terminal group bonded to the dimethylene radical can be, for example, -OH, and the terminal group bonded to the oxygen can be, for example, hydrogen. The terminal groups can also be polyether radicals.

In the compounds of the formula (B-10), the end group bonded to the -CH₂- residue can be, for example, hydrogen and the end group bonded to the -CH(CO₂A₇) residue can be, for example, -CH=CH-COOA₇.

 E_1 , E_8 , E_{12} , E_{13} , E_{16} , E_{18} , E_{22} , E_{23} , E_{25} , E_{29} , R_6 , R_{13} , R_{16} , R_{18} , R_{30} and R_{32} are preferably hydrogen, C_1 - C_4 alkyl, C_1 - C_{10} alkoxy, cyclohexyloxy, allyl, benzyl or acetyl.

 E_1 , E_8 , E_{12} , E_{13} , E_{16} , E_{18} , E_{22} , E_{23} , E_{25} , E_{29} , R_6 , R_{13} , R_{16} , R_{18} , R_{30} and R_{32} are in particular hydrogen or methyl and E_1 and R_6 additionally are C_1 - C_8 alkoxy.

According to a preferred embodiment,

m₁ is 1, 2 or 4,

if m₁ is 1, E₂ is C₁₂-C₂₀alkyl,

if m₁ is 2, E₂ is C₂-C₁₀alkylene or a group of the formula (a-1)

E₃ is C₁-C₄alkyl,

E4 is C1-C6alkylene, and

E₅ and E₆ independently of one another are C₁-C₄alkyl, and

if m₁ is 4, E₂ is C₄-C₈alkanetetrayl;

two of the radicals E7 are -COO-(C10-C15alkyl), and

two of the radicals E₇ are a group of the formula (a-II);

E₉ and E₁₀ together form C₉-C₁₃alkylene,

E₁₁ is hydrogen or a group -Z₁-COO-Z₂,

Z₁ is C₂-C₆alkylene, and

Z₂ is C₁₀-C₁₆alkyl;

E₁₄ is hydrogen, and

E₁₅ is C₂-C₆alkylene or C₃-C₅alkylidene;

E₁₇ is C₁₀-C₁₄alkyl;

E24 is C1-C4alkoxy;

m₂ is 1, 2 or 3,

when m_2 is 1, E_{26} is a group — $CH_2CH_2-NH-\langle \rangle$

when m₂ is 2, E₂₆ is C₂-C₆alkylene, and

when m₂ is 3, E₂₆ is a group of the formula (a-IV)

the radicals E27 independently of one another are C2-C6alkylene, and

the radicals E_{28} independently of one another are $C_1\text{-}C_4$ alkyl or $C_5\text{-}C_8$ cycloalkyl; and

E₃₀ is C₂-C₈alkylene;

R₁ and R₃ independently of one another are a group of the formula (b-I),

R2 is C2-CBalkylene,

 R_4 and R_5 independently of one another are hydrogen, C_1 - C_{12} alkyl, C_5 - C_8 cycloalkyl or a group of the formula (b-l), or the radicals R_4 and R_5 , together with the nitrogen atom to which they are bonded, form a 5- to 10-membered heterocyclic ring, and

b₁ is a number from 2 to 25;

R₇ and R₁₁ independently of one another are hydrogen or C₁-C₄alkyl,

R₈, R₉ and R₁₀ independently of one another are C₂-C₄alkylene, and

 X_1 , X_2 , X_3 , X_4 , X_5 , X_6 , X_7 and X_8 independently of one another are a group of the formula (b-II).

R₁₂ is hydrogen, C₁-C₄alkyl, C₅-C₈cycloalkyl or a group of the formula (b-l);

R₁₄ is C₁-C₄alkyl,

R₁₅ is C₃-C₆alkylene, and

b₂ is a number from 2 to 25;

R₁₇ and R₂₁ independently of one another are a direct bond or a group

 $-N(X_9)-CO-X_{10}-CO-N(X_{11})-$

X₉ and X₁₁ independently of one another are hydrogen or C₁-C₄alkyl,

X₁₀ is a direct bond,

R₁₉ and R₂₃ are C₁-C₂₅alkyl or phenyl,

R₂₀ and R₂₄ are hydrogen or C₁-C₄aikyl,

R₂₂ is C₁-C₂₅alkyl or a group of the formula (b-l), and

b₃ is a number from 1 to 25;

R₂₅, R₂₆, R₂₇, R₂₈ and R₂₉ independently of one another are a direct bond or

C1-C4alkylene, and

b₄ is a number from 1 to 25;

b'5, b"5 and b"5 independently of one another are a number from 2 to 4, and

R₃₁ is hydrogen, C₁-C₄alkyl, C₅-C₈cycloalkyl, phenyl or benzyl;

A₁ is hydrogen or methyl,

A2 is a direct bond or C2-C6alkylene, and

 n_1 is a number from 2 to 25;

n₂ and n₂* are a number from 2 to 25;

A₃ and A₄ independently of one another are hydrogen or C₁-C₄alkyl, or A₃ and A₄ together

form a C9-C13alkylene group, and

the variables n₃ independently of one another are a number from 1 to 25;

n₄ is a number from 2 to 25,

 A_5 and A_6 independently of one another are C_1 - C_4 alkyl, and A_7 is C_1 - C_4 alkyl or a group of the formula (b-l) with the proviso that at least 50 % of the radicals A_7 are a group of the formula (b-l).

The two different sterically hindered amine compounds of component (I) are preferably selected from the group consisting of the compounds of the formulae (A-1-a), (A-1-b), (A-1-c), (A-1-d), (A-2-a), (A-3-a), (A-3-b), (A-4-a), (A-4-b), (A-5), (A-6-a), (A-7), (A-8-a), (A-9-a), (A-9-b), (A-9-c), (A-10-a), (B-1-a), (B-1-b), (B-1-c), (B-1-d), (B-2-a), (B-3-a), (B-4-a), (B-4-b) and (B-4-c), a product (B-6-a) and the compounds of the formulae (B-7-a), (B-8-a), (B-8-b), (B-9-a) and (B-10-a);

$$H_3C$$
 CH_3
 CH_3
 CCH_3
 CCH_3

$$\begin{bmatrix} H_3C & CH_3 & O \\ E_1 - N & O - C \end{bmatrix} \xrightarrow{C_4H_9} CH_2 CH_2$$

$$C = CH_2 CH_3$$

$$C(CH_3)_3$$

$$C(CH_3)_3$$

$$C(CH_3)_3$$

wherein E_1 is hydrogen, C_1 - C_8 alkyl, O-, -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl;

in which two of the radicals E7 are -COO-C13H27 and

two of the radicals E_7 are $COO \longrightarrow N \longrightarrow E_8$ and E_8 has one of the meanings $H_3C \longrightarrow CH_3$

of E1;

$$(CH2)9 O C = O$$

$$H3C O CH3$$

$$CH3 CH3$$

$$CH3 CH3$$

$$(CH_{2})_{9} CH_{2}CH_{2}CC-O-(C_{12}-C_{14}alkyt)$$

$$CH_{2}OC-O$$

$$H_{3}CC+CH_{3}CH_{3}CH_{3}$$

$$CH_{3}CH_{3}CH_{3}$$

$$CH_{3}CH_{3}$$

wherein E₁₂ has one of the meanings of E₁;

wherein E_{13} has one of the meanings of E_1 ;

$$H_{3}C CH_{3}$$

$$H_{3}C CH_{3}$$

$$N H_{3}C CH_{3}$$

$$H_{3}C CH_{3}$$

$$H_{3}C CH_{3}$$

$$H_{3}C CH_{3}$$

$$H_{3}C CH_{3}$$

wherein E₁₆ has one of the meanings of E_{1;}

$$H_{25}C_{12}$$
 N
 H_3C
 CH_3
 CH_3
 CH_3
 O
 H_3C
 CH_3

wherein E_{18} has one of the meanings of $E_{1;}$

$$E_{10} \longrightarrow E_{21}$$

$$E_{20} \longrightarrow E_{20}$$

$$(A-7)$$

in which E_{19} , E_{20} and E_{21} independently of one another are a group of the formula (a-III)

wherein E22 has one of the meanings of E1;

$$H_{3}C CH_{3}$$

wherein E23 has one of the meanings of E1;

wherein E25 has one of the meanings of E1;

wherein E29 has one of the meanings of E1;

wherein b_1 is a number from 2 to 20 and R_6 is hydrogen, C_1 - C_8 alkyl, O_7 - OH_7 - OH_7 - OH_7 - OH_7 - OH_8

wherein R₁₃ has one of the meanings of R₆,

(B-3-a)

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wherein b_2 is a number from 2 to 20 and R_{16} has one of the meanings of R_6 ;

wherein b_3 is a number from 1 to 20 and R_{18} has one of the meanings of R_{6} ;

wherein b4 is a number from 1 to 20 and R30 has one of the meanings of R6;

a product (B-6-a) obtainable by reacting a product, obtained by reaction of a polyamine of the formula (B-6-1-a) with cyanuric chloride, with a compound of the formula (B-6-2-a)

$$H_2N - (CH_2) - NH - (CH_2) - NH - (CH_2) - NH_2$$
 (B-6-1-a)

$$H_3C$$
 H_3C
 CH_3
 CH_3
 CH_3

in which R₃₂ has one of the meanings of R₆.

wherein n₁ is a number from 2 to 20;

$$\begin{array}{c|c}
CH - CH_2 - O \\
CH_2 \\
CH_2 \\
CH_2 \\
CH_3 \\
CH_4 \\
CH_5 \\
CH_5$$

wherein n₂ and n₂* are a number from 2 to 20;

(B-9-a)

wherein the variables n₃ independently of one another are a number from 1 to 20;

wherein n_4 is a number from 2 to 20, and at least 50 % of the radicals A_7 are a group of the formula (b-I)

wherein R_6 is hydrogen, C_1 - C_8 alkyl, O° , -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl, and the remaining radicals A_7 are ethyl.

According to a preferred embodiment,

the two different sterically hindered amine compounds of component (I) are

- 1) a compound of the formula (A-1-b) wherein E₁ is hydrogen, and a compound of the formula (B-1-a) wherein R₆ is hydrogen;
- 2) a compound of the formula (B-1-a) wherein R_6 is hydrogen, and a compound of the formula (B-7-a); or
- 3) a compound of the formula (B-2-a) wherein R₁₃ is methyl, and a compound of the formula (B-7-a).

The organic salt of zinc or magnesium defined in component (II) is preferably a compound of the formula MeL_2 in which Me is zinc or magnesium and L is an anion of an organic acid or of an enol. The organic acid can, for example, be a sulfonic acid, sulfinic acid, phosphonic acid or phosphinic acid, but is preferably a carboxylic acid. The acid can be aliphatic, aromatic, araliphatic or cycloaliphatic; it can be linear or branched; it can be substituted by

hydroxyl or alkoxy groups; it can be saturated or unsaturated and it preferably contains 1 to 24 carbon atoms.

Examples of carboxylic acids of this type are formic, acetic, propionic, butyric, isobutyric, caprioic, 2-ethylcaproic, caprylic, capric, lauric, palmitic, stearic, behenic, oleic, lactic, ricinoleic, 2-ethoxypropionic, benzoic, salicylic, 4-butylbenzoic, toluic, 4-dodecylbenzoic, phenylacetic, naphthylacetic, cyclohexanecarboxylic, 4-butylcyclohexanecarboxylic or cyclohexylacetic acid. The carboxylic acid can also be a technical mixture of carboxylic acids, for example technical mixtures of fatty acids or mixtures of alkylated benzoic acids.

Examples of organic acids containing sulfur or phosphorus are methanesulfonic, ethanesulfonic, α,α -dimethylethanesulfonic, n-butanesulfonic, n-dodecanesulfonic, benzenesulfonic, toluenesulfonic, 4-nonylbenzenesulfonic, 4-dodecylbenzenesulfonic or cyclohexanesulfonic acid, dodecanesulfinic, benzenesulfinic or naphthalenesulfinic acid, butylphosphonic acid, phenylphosphonic acid, monomethyl or monoethyl phenylphosphonate, monobutyl benzylphosphonate, dibutylphosphinic acid or diphenylphosphinic acid.

If L is an enolate anion, it is preferably an anion of a β -dicarbonyl compound or of an o-acylphenol. Examples of β -dicarbonyl compounds are acetylacetone, benzoylacetone, dibenzoylmethane, ethyl acetoacetate, butyl acetoacetate, lauryl acetoacetate or α -acetylcyclohexanone. Examples of o-acylphenols are 2-acetylphenol, 2-butyroylphenol, 2-acetyl-1-naphthol, 2-benzoylphenol or salicylaldehyde. The enolate is preferably the anion of a β -dicarbonyl compound having 5 to 20 carbon atoms.

Organic salts of zinc or magnesium are preferably an acetylacetonate or an aliphatic monocarboxylate having, for example, 1 to 24 carbon atoms. Magnesium acetate, laurate and stearate, zinc formate, acetate, oenanthate, laurate and stearate as well as zinc acetylacetonate and magnesium acetylacetonate are some of the particular preferred examples.

Zinc stearate, magnesium stearate, zinc acetylacetonate, magnesium acetylacetonate, zinc acetate and magnesium acetate are of special interest.

The inorganic salt of zinc or magnesium is for example a carbonate containing compound such as

- Zn-hydroxide-carbonate, Mg-hydroxide-carbonate, dolomite, e.g a Ca/Mg carbonate such as Microdol Super (RTM) from Micro Minerals (RTM); or
- a natural or synthetic hydrotalcite.

The natural hydrotalcite is held to possess a structure Mg₆Al₂(OH)₁₆CO₃ · 4 H₂O. A typical empirical formula of a synthetic hydrotalcite is

Al₂Mg_{4.35}OH_{11.36}CO_{3(1.67)} • x H₂O.

Examples of the synthetic product include:

 $Mg_{0.7}Al_{0.3}(OH)_2(CO_3)_{0.15} \cdot 0.54 H_2O_1$

 $Mg_{4,5}Al_2(OH)_{13}CO_3 \cdot 3.5 H_2O$, or

Mg_{4,2}AI(OH)_{12,4}CO₃.

Preferred synthetic hydrotalcites are L-55R II (RTM) from REHEIS (RTM) as well as ZHT-4A (RTM) and DHT-4A (RTM) from Kyowa Chemical Industry Co (RTM).

Component (II) can also be a mixture of two different Mg- and/or Zn- compounds, for example

- Mg-stearate and hydrotalcite (DHT-4A (RTM)),
- Zn-stearate and hydrotalcite (DHT-4A (RTM)),
- Mg-acetylacetonate and hydrotalcite (DHT-4A (RTM)),
- Mg-oxide and hydrotalcite (DHT-4A (RTM)),
- Mg-hydroxide and hydrotalcite (DHT-4A (RTM)),
- Zn-hydroxide-carbonate and Mg-stearate,
- Zn-hydroxide-carbonate and Zn-stearate,
- Zn-hydroxide-carbonate and Mg-acetylacetonate,
- Zn-hydroxide-carbonate and Mg-oxide,
- Zn-hydroxide-carbonate and Zn-oxide,
- Zn-hydroxide-carbonate and Mg-hydroxide,
- hydrotalcite (REHEIS (RTM)) and Mg-stearate,
- hydrotalcite (REHEIS (RTM)) and Zn-stearate,

- hydrotalcite (REHEIS (RTM)) and Mg-oxide,
- dolomite (Microdol Super (RTM)) and Zn-stearate,
- dolomite (Microdol Super (RTM)) and Mg-stearate,
- dolomite (Microdol Super (RTM)) and Zn-oxide,
- dolomite (Microdol Super (RTM)) and Mg-hydroxide,
- Mg-stearate and Zn-stearate,
- Mg-stearate and Zn-acetylacetonate,
- Mg-stearate and Mg-oxide,
- Mg-stearate and Zn-oxide,
- · Mg-stearate and Mg-hydroxide,
- Zn-stearate and Mg-acetate,
- Zn-stearate and Mg-oxide,
- Zn-stearate and Mg-hydroxide,
- Mg-acetylacetonate and Zn-acetylacetonate,
- Mg-acetylacetonate and Mg-oxide,
- Mg-acetylacetonate and Zn-oxide,
- Mg-acetylacetonate and Mg-hydroxide,
- Zn-acetylacetonate and Mg-oxide,
- Zn-acetylacetonate and Zn-oxide, or
- Mg-oxide and Zn-oxide.

In this case, the two different compounds of component (II) may be present in a weight ratio of 1:10 to 10:1.

A preferred embodiment of this invention relates to a stabilizer mixture wherein the compound(s) of component (II) is (are) selected from the group consisting of Mg carboxylates, Zn carboxylates, Mg oxides, Zn oxides, Mg hydroxides, Zn hydroxides, Mg carbonates and Zn carbonates.

Another preferred embodiment of this invention relates to a stabilizer mixture wherein component (II) is a Mg carboxylate, a Zn carboxylate, a hydrotalcite, or a mixture of a Mg carboxylate and a hydrotalcite.

A particularly preferred embodiment of this invention relates to a stabilizer mixture wherein component (II) is Mg stearate, Zn stearate, a hydrotalcite, in particular DHT-4A (RTM), or a mixture of Mg stearate and a hydrotalcite.

Preferred examples of stabilizer mixtures according to the present invention are:

- 1. TINUVIN 622 (RTM) + CHIMASSORB 944 (RTM) + Mg stearate
- 2. TINUVIN 770 (RTM) + CHIMASSORB 944 (RTM) + Mg stearate
- 3. TINUVIN 622 (RTM) + CHIMASSORB 119 (RTM) + Mg stearate
- 4. TINUVIN 770 (RTM) + UVASORB HA 88 (RTM) + Mg stearate
- 5. TINUVIN 622 (RTM) + UVASORB HA 88 (RTM) + Mg stearate
- 6. TINUVIN 622 (RTM) + UVASIL 299 (RTM) + Mg stearate
- 7. TINUVIN 622 (RTM) + CHIMASSORB 2020 (RTM) + Mg stearate
- 8. TINUVIN 770 (RTM) + CHIMASSORB 2020 (RTM) + Mg stearate
- 9. TINUVIN 622 (RTM) + CHIMASSORB 944 (RTM) + DHT-4A (RTM)
- 10.TINUVIN 770 (RTM) + CHIMASSORB 944 (RTM) + DHT-4A (RTM)
- 11.TINUVIN 622 (RTM) + CHIMASSORB 119 (RTM) + DHT-4A (RTM)
- 12.TINUVIN 770 (RTM) + UVASORB HA 88 (RTM) + DHT-4A (RTM)
- 13.TINUVIN 622 (RTM) + UVASORB HA 88 (RTM) + DHT-4A (RTM)
- 14.TINUVIN 622 (RTM) + UVASIL 299 (RTM) + DHT-4A (RTM)
- 15.TINUVIN 622 (RTM) + CHIMASSORB 2020 (RTM) + DHT-4A (RTM)
- 16.TINUVIN 770 (RTM) + CHIMASSORB 2020 (RTM) + DHT-4A (RTM)
- 17.TINUVIN 622 (RTM) + CHIMASSORB 944 (RTM) + DHT-4A (RTM) + Mg stearate
- 18.TINUVIN 770 (RTM) + CHIMASSORB 944 (RTM) + DHT-4A (RTM) + Mg stearate
- 19.TINUVIN 622 (RTM) + CHIMASSORB 119 (RTM) + DHT-4A (RTM) + Mg stearate
- 20.TINUVIN 770 (RTM) + UVASORB HA 88 (RTM) + DHT-4A (RTM) + Mg stearate
- 21.TINUVIN 622 (RTM) + UVASORB HA 88 (RTM) + DHT-4A (RTM) + Mg stearate
- 22.TINUVIN 622 (RTM) + UVASIL 299 (RTM) + DHT-4A (RTM) + Mg stearate
- 23.TINUVIN 622 (RTM) + CHIMASSORB 2020 (RTM) + DHT-4A (RTM) + Mg stearate
- 24.TINUVIN 770 (RTM) + CHIMASSORB 2020 (RTM) + DHT-4A (RTM) + Mg stearate

The commercial product TINUVIN 622 (RTM) corresponds to the compound of the formula (B-7-a).

The commercial product CHIMASSORB 944 (RTM) corresponds to the compound of the formula (B-1-a) wherein R_6 is hydrogen.

The commercial product TINUVIN 770 (RTM) corresponds to the compound of the formula (A-1-b) wherein E₁ is hydrogen.

The commercial product CHIMASSORB 119 (RTM) corresponds to the compound of the formula (B-2-a) wherein R₁₃ is methyl.

The commercial product UVASORB HA 88 (RTM) corresponds to the product (B-6-a) wherein R₃₂ is hydrogen.

The commercial product UVASIL 299 (RTM) corresponds to the compound of the formula (B-3-a) wherein R_{16} is hydrogen.

The commercial product CHIMASSORB 2020 (RTM) corresponds to the compound of the formula (B-1-b) wherein R_6 is hydrogen.

A further preferred embodiment of this invention relates to a stabilizer mixture containing additionally

(X-1) a pigment or

(X-2) an UV absorber or

(X-3) a pigment and an UV absorber.

The pigment (component (X-1)) may be an inorganic or organic pigment.

Examples of inorganic pigments are titanium dioxide, zinc oxide, carbon black, cadmium sulfide, cadmium selenide, chromium oxide, iron oxide, lead oxide and so on.

Examples of organic pigments are azo pigments, anthraquinones, phthalocyanines, tetrachloroisoindolinones, quinacridones, isoindolines, perylenes, pyrrolopyrroles (such as Pigment Red 254) and so on.

All pigments described in "Gächter/Müller: Plastics Additives Handbook, 3rd Edition, Hanser Publishers, Munich Vienna New York", page 647 to 659, point 11.2.1.1 to 11.2.4.2 can be used as component (X-1).

A particularly preferred pigment is titanium dioxide, optionally in combination with an organic pigment.

Examples of such organic pigments are:

C.I. (Colour Index) Pigment Yellow 93, C.I. Pigment Yellow 95, C.I. Pigment Yellow 138, C.I. Pigment Yellow 139, C.I. Pigment Yellow 155, C.I. Pigment Yellow 162, C.I. Pigment Yellow 168, C.I. Pigment Yellow 180, C.I. Pigment Yellow 183, C.I. Pigment Red 44, C.I. Pigment Red 170, C.I. Pigment Red 202, C.I. Pigment Red 214, C.I. Pigment Red 254, C.I. Pigment Red 264, C.I. Pigment Red 272, C.I. Pigment Red 48:2, C.I. Pigment Red 48:3, C.I. Pigment Red 53:1, C.I. Pigment Red 57:1, C.I. Pigment Green 7, C.I. Pigment Blue 15:1, C.I. Pigment Blue 15:3 and C.I. Pigment Violet 19.

Examples of the UV absorber (component (X-2)) are a 2-(2'-hydroxyphenyl)benzotriazole, a 2-hydroxybenzophenone, an ester of substituted or unsubstituted benzoic acid, an acrylate, an oxamide, a 2-(2-hydroxyphenyl)-1,3,5-triazine, a monobenzoate of resorcinol or a formamidine.

The 2-(2'-hydroxyphenyl)benzotriazole is e.g. 2-(2'-hydroxy-5'-methylphenyl)-benzotriazole. 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl- 2'-hydroxy-5'-methylphenyl)-5chloro-benzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis-(α,α-dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, mixture of 2-(3'-tert-butyl-2'hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-5'-[2-(2ethylhexyloxy)-carbonylethyl]-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'hydroxy-5'-(2-methoxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2octyloxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenylbenzotriazole, 2,2'-methylene-bis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazole-2-ylphenol] or the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotri-

azole with polyethylene glycol 300; $[R-CH_2CH_2-COO(CH_2)_3-]_2$ where R=3'-tert-butyl-4'-hydroxy-5'-2H-benzotriazol-2-ylphenyl.

2-(3',5'-Di-tert-butyl-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chloro-benzotriazole and 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)-benzotriazole are preferred.

The 2-hydroxybenzophenone is for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy or 2'-hydroxy-4,4'-dimethoxy derivatives.

2-Hydroxy-4-octyloxybenzophenone is preferred.

The ester of a substituted or unsubstituted benzoic acid is for example 4-tert-butyl-phenyl salicylate, phenyl salicylate, octylphenyl salicylate, dibenzoyl resorcinol, bis(4-tert-butylbenzoyl) resorcinol, benzoyl resorcinol, 2,4-di-tertbutylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate, hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, octadecyl 3,5-di-tert-butyl-4-hydroxybenzoate.

2,4-Di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate and hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate are preferred.

The acrylate is for example ethyl α -cyano- β , β -diphenylacrylate, isooctyl α -cyano- β , β -diphenylacrylate, methyl α -carbomethoxycinnamate, methyl α -cyano- β -methyl-p-methoxycinnamate, butyl α -cyano- β -methyl-p-methoxy-cinnamate, methyl α -carbomethoxy-p-methoxycinnamate or N-(β -carbomethoxy- β -cyanovinyl)-2-methylindoline.

The oxamide is for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide or its mixture with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide or mixtures of ortho- and para-methoxy-disubstituted oxanilides.

The 2-(2-hydroxyphenyl)-1,3,5-triazine is for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-

(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-octyloxy-propyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxy-phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxy-propoxy)phenyl]-1,3,5-triazine or 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine.

2-(2-Hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine and 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine are preferred.

The monobenzoate of resorcinol is for example the compound of the formula

The formamidine is for example the compound of the formula

$$H_5C_2O$$
 C
 N
 C_2H_5

The UV absorber is in particular a

2-(2'-hydroxyphenyl)benzotriazole, a 2-hydroxybenzophenone or a hydroxyphenyltriazine.

A further preferred embodiment of this invention relates to a stabilizer mixture which additionally contains as a further component (XX) an organic salt of Ca, an inorganic salt of Ca, Ca oxide or Ca hydroxide.

Examples of an organic salt of Ca are Ca-stearate, Ca-laurate, Ca-lactate and Ca-stearoyllactate.

Examples of an inorganic salt of Ca are CaCO₃, CaCl₂, CaF₂, Ca₃(PO₄)₂, CaHPO₄, Ca(PO₃)₂, Ca₂P₂O₇, CaSO₄ and CaSio₃.

Further preferred examples of stabilizer mixtures according to the present invention are:

- a. TINUVIN 622 (RTM) + CHIMASSORB 944 (RTM) + Mg stearate + Ca stearate
- b. TINUVIN 770 (RTM) + CHIMASSORB 944 (RTM) + Mg stearate + Ca stearate
- c. TINUVIN 622 (RTM) + CHIMASSORB 119 (RTM) + Mg stearate + Ca stearate
- d. TINUVIN 770 (RTM) + UVASORB HA 88 (RTM) + Mg stearate + Ca stearate
- e. TINUVIN 622 (RTM) + UVASORB HA 88 (RTM) + Mg stearate + Ca stearate
- f. TINUVIN 622 (RTM) + UVASIL 299 (RTM) + Mg stearate + Ca stearate
- g. TINUVIN 622 (RTM) + CHIMASSORB 2020 (RTM) + Mg stearate + Ca stearate
- h. TINUVIN 770 (RTM) + CHIMASSORB 2020 (RTM) + Mg stearate + Ca stearate
- i. TINUVIN 622 (RTM) + CHIMASSORB 944 (RTM) + DHT-4A (RTM) + Ca stearate
- j. TINUVIN 770 (RTM) + CHIMASSORB 944 (RTM) + DHT-4A (RTM) + Ca stearate
- k. TINUVIN 622 (RTM) + CHIMASSORB 119 (RTM) + DHT-4A (RTM) + Ca stearate
- I. TINUVIN 770 (RTM) + UVASORB HA 88 (RTM) + DHT-4A (RTM) + Ca stearate
- m. TINUVIN 622 (RTM) + UVASORB HA 88 (RTM) + DHT-4A (RTM) + Ca stearate
- n. TINUVIN 622 (RTM) + UVASIL 299 (RTM) + DHT-4A (RTM) + Ca stearate
- o. TINUVIN 622 (RTM) + CHIMASSORB 2020 (RTM) + DHT-4A (RTM) + Ca stearate
- p. TINUVIN 770 (RTM) + CHIMASSORB 2020 (RTM) + DHT-4A (RTM) + Ca stearate

The stabilizer mixture according to this invention is suitable for stabilizing organic materials against degradation induced by light, heat or oxidation. Examples of such organic materials are the following:

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optionally can be crosslinked), for example high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), (VLDPE) and (ULDPE), or polyvinyl cyclohexane.

Polyolefins, i.e. the polymers of monoolefins exemplified in the preceding paragraph, preferably polyethylene and polypropylene, can be prepared by different, and especially by the following, methods:

- a) radical polymerisation (normally under high pressure and at elevated temperature).
- b) catalytic polymerisation using a catalyst that normally contains one or more than one metal of groups IVb, Vb, Vlb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls that may be either π- or σ-coordinated. These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups Ia, IIa and/or IIIa of the Periodic Table. The activators may be modified conveniently with further ester, ether, amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler (-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).
- 2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE).

- 3. Copolymers of monoolefins and diolefins with each other or with other vinyl monomers, for example ethylene/propylene copolymers, linear low density polyethylene (LLDPE) and mixtures thereof with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers, ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers and their copolymers with carbon monoxide or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with propylene and a diene such as hexadiene, dicyclopentadiene or ethylidene-norbornene; and mixtures of such copolymers with one another and with polymers mentioned in 1) above, for example polypropylene/ethylene-propylene copolymers, LDPE/ethylene-vinyl acetate copolymers (EVA), LDPE/ethylene-acrylic acid copolymers (EAA), LLDPE/EVA, LLDPE/EAA and alternating or random polyalkylene/carbon monoxide copolymers and mixtures thereof with other polymers, for example polyamides.
- 4. Hydrocarbon resins (for example C_5 - C_9) including hydrogenated modifications thereof (e.g. tackifiers) and mixtures of polyalkylenes and starch.
- 5. Polystyrene, poly(p-methylstyrene), poly(α -methylstyrene).
- 6. Copolymers of styrene or α-methylstyrene with dienes or acrylic derivatives, for example styrene/butadiene, styrene/acrylonitrile, styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methyl acrylate; mixtures of high impact strength of styrene copolymers and another polymer, for example a polyacrylate, a diene polymer or an ethylene/propylene/diene terpolymer; and block copolymers of styrene such as styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/ styrene.
- 7. Graft copolymers of styrene or α-methylstyrene, for example styrene on polybutadiene, styrene on polybutadiene-styrene or polybutadiene-acrylonitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) on polybutadiene; styrene, acrylonitrile and methyl methacrylate on polybutadiene; styrene and maleic anhydride on polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide on polybutadiene; styrene and maleimide on polybutadiene; styrene and maleimide on polybutadiene;

butadiene; styrene and alkyl acrylates or methacrylates on polybutadiene; styrene and acrylonitrile on ethylene/propylene/diene terpolymers; styrene and acrylonitrile on polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile on acrylate/butadiene copolymers, as well as mixtures thereof with the copolymers listed under 6), for example the copolymer mixtures known as ABS, MBS, ASA or AES polymers.

- 8. Halogen-containing polymers such as polychloroprene, chlorinated rubbers, chlorinated and brominated copolymer of isobutylene-isoprene (halobutyl rubber), chlorinated or sulfo-chlorinated polyethylene, copolymers of ethylene and chlorinated ethylene, epichlorohydrin homo- and copolymers, especially polymers of halogen-containing vinyl compounds, for example polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride, polyvinylidene fluoride, as well as copolymers thereof such as vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate copolymers.
- 9. Polymers derived from α,β -unsaturated acids and derivatives thereof such as polyacrylates and polymethacrylates; polymethyl methacrylates, polyacrylamides and polyacrylonitriles, impact-modified with butyl acrylate.
- 10. Copolymers of the monomers mentioned under 9) with each other or with other unsaturated monomers, for example acrylonitrile/ butadiene copolymers, acrylonitrile/alkyl acrylate copolymers, acrylonitrile/alkoxyalkyl acrylate or acrylonitrile/vinyl halide copolymers or acrylonitrile/ alkyl methacrylate/butadiene terpolymers.
- 11. Polymers derived from unsaturated alcohols and amines or the acyl derivatives or acetals thereof, for example polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallyl melamine; as well as their copolymers with olefins mentioned in 1) above.
- 12. Homopolymers and copolymers of cyclic ethers such as polyalkylene glycols, polyethylene oxide, polypropylene oxide or copolymers thereof with bisglycidyl ethers.
- 13. Polyacetals such as polyoxymethylene and those polyoxymethylenes which contain ethylene oxide as a comonomer; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.

- 14. Polyphenylene oxides and sulfides, and mixtures of polyphenylene oxides with styrene polymers or polyamides.
- 15. Polyurethanes derived from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as precursors thereof.
- 16. Polyamides and copolyamides derived from diamines and dicarboxylic acids and/or from aminocarboxylic acids or the corresponding lactams, for example polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides starting from m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic or/and terephthalic acid and with or without an elastomer as modifier, for example poly-2,4,4,-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide; and also block copolymers of the aforementioned polyamides with polyolefins, olefin copolymers, ionomers or chemically bonded or grafted elastomers; or with polyethers, e.g. with polyethylene glycol, polypropylene glycol or polytetramethylene glycol; as well as polyamides or copolyamides modified with EPDM or ABS; and polyamides condensed during processing (RIM polyamide systems).
- 17. Polyureas, polyimides, polyamide-imides, polyetherimids, polyesterimids, polyhydantoins and polybenzimidazoles.
- 18. Polyesters derived from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or the corresponding lactones, for example polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate, polyalkylene naphthalate (PAN) and polyhydroxybenzoates, as well as block copolyether esters derived from hydroxyl-terminated polyethers; and also polyesters modified with polycarbonates or MBS.
- 19. Polycarbonates and polyester carbonates.
- 20. Polysulfones, polyether sulfones and polyether ketones.

- 21. Crosslinked polymers derived from aldehydes on the one hand and phenols, ureas and melamines on the other hand, such as phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.
- 22. Drying and non-drying alkyd resins.
- 23. Unsaturated polyester resins derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols and vinyl compounds as crosslinking agents, and also halogen-containing modifications thereof of low flammability.
- 24. Crosslinkable acrylic resins derived from substituted acrylates, for example epoxy acrylates, urethane acrylates or polyester acrylates.
- 25. Alkyd resins, polyester resins and acrylate resins crosslinked with melamine resins, urea resins, isocyanates, isocyanurates, polyisocyanates or epoxy resins.
- 26. Crosslinked epoxy resins derived from aliphatic, cycloaliphatic, heterocyclic or aromatic glycidyl compounds, e.g. products of diglycidyl ethers of bisphenol A and bisphenol F, which are crosslinked with customary hardeners such as anhydrides or amines, with or without accelerators.
- 27. Natural polymers such as cellulose, rubber, gelatin and chemically modified homologous derivatives thereof, for example cellulose acetates, cellulose propionates and cellulose butyrates, or the cellulose ethers such as methyl cellulose; as well as rosins and their derivatives.
- 28. Blends of the aforementioned polymers (polyblends), for example PP/EPDM, Polyamide/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR, POM/acrylate, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO, PBT/PC/ABS or PBT/PET/PC.
- 29. Naturally occurring and synthetic organic materials which are pure monomeric compounds or mixtures of such compounds, for example mineral oils, animal and vegetable fats,

oil and waxes, or oils, fats and waxes based on synthetic esters (e.g. phthalates, adipates, phosphates or trimellitates) and also mixtures of synthetic esters with mineral oils in any weight ratios, typically those used as spinning compositions, as well as aqueous emulsions of such materials.

30. Aqueous emulsions of natural or synthetic rubber, e.g. natural latex or latices of carboxylated styrene/butadiene copolymers.

This invention therefore additionally relates to a composition comprising an organic material subject to degradation induced by light, heat or oxidation and the stabilizer mixture described above.

A further embodiment of the present invention is a method for stabilizing an organic material against degradation induced by light, heat or oxidation, which comprises incorporating into the organic material the stabilizer mixture described above.

The organic material is preferably a synthetic polymer, in particular from one of the above groups. Polyolefins are preferred and polyethylene, polypropylene, a polyethylene copolymer or a polypropylene copolymer are particularly preferred.

The components (I), (II) and optionally (X-1) and/or (X-2), as well as optionally component (XX) may be added to the organic material to be stabilized either individually or mixed with one another.

The total amount of the two different sterically hindered amine compounds (component (I)) in the organic material to be stabilized is preferably 0.005 to 5 %, in particular 0.01 to 1 % or 0.05 to 1 %, relative to the weight of the organic material.

The Mg and/or Zn compounds (component (II)) are present in the organic material in a total amount of preferably 0.005 to 1 %, in particular 0.05 to 0.2 %, relative to the weight of the organic material.

The pigment (component (X-1)) is optionally present in the organic material in an amount of preferably 0.01 to 10 %, in particular 0.05 to 1 %, relative to the weight of the organic material.

The UV absorber (component (X-2)) is optionally present in the organic material in an amount of preferably 0.01 to 1 %, in particular 0.05 to 0.5 %, relative to the weight of the organic material.

The total amount of component (X-3) (the pigment in combination with the UV absorber) is preferably 0.01 to 10 %, relative to the weight of the organic material. The weight ratio of the UV absorber to the pigment is for example 2:1 to 1:10.

When the pigment used is titanium dioxide in combination with an organic pigment as described above, titanium dioxide is preferably present in the organic material in an amount of 0.01 to 5 %, relative to the weight of the organic material, and the organic pigment may be present in an amount of, for example, 0.01 to 2 %, relative to the weight of the organic material.

The Ca compound (component (XX)) is optionally present in the organic material in an amount of e.g. 0.005 to 1 %, preferably 0.05 to 0.2 %.

The weight ratio of the two different sterically hindered amine compounds forming component (I) is for example 1:10 to 10:1, preferably 1:5 to 5:1, in particular 1:2 to 2:1.

The weight ratio of the components (I):(II) is for example 1:10 to 20:1, preferably 1:5 to 5:1, in particular 1:2 to 2:1.

The weight ratio of the components (I):(X-1) is for example 1:10 to 10:1, preferably 1:5 to 5:1, in particular 1:2 to 2:1.

The weight ratio of the components (I):(X-2) is for example 1:5 to 5:1, preferably 1:2 to 2:1.

The weight ratio of the components (I):(X-3) is for example 1:10 to 10:1, preferably 1:5 to 5:1, in particular 1:2 to 2:1.

The weight ratio of the components (I):(XX) is for example 1:10 to 10:1, preferably 1:5 to 5:1, in particular 1:2 to 2:1.

The above components can be incorporated into the organic material to be stabilized by known methods, for example before or during shaping or by applying the dissolved or dispersed compounds to the organic material, if necessary with subsequent evaporation of the solvent. The components can be added to the organic material in the form of a powder, granules or a masterbatch, which contains these components in, for example, a concentration of from 2.5 to 25% by weight.

If desired, the components (I), (II) and optionally (X-1) and/or (X-2), as well as component (XX) can be melt blended with each other before incorporation in the organic material. They can be added to a polymer before or during the polymerization or before the crosslinking.

The materials stabilized according to this invention can be used in a wide variety of forms, for example as films, fibres, tapes, moulding compositions, profiles or as binders for paints, adhesives or putties.

The stabilized material may additionally also contain various conventional additives, for example :

1. Antioxidants

1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butyl-phenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-(α-methylcyclohexyl)-4,6-dimethylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tri-cyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, nonylphenols which are linear or branched in the side chains, for example, 2,6-di-nonyl-4-methyl-phenol, 2,4-dimethyl-6-(1'-methyl-dimethyl-dimethyl-6-(1'-methyl-dimethyl-dimethyl-dimethyl-6-(1'-methyl-dimethyl-

heptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.

- 1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.
- 1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl) adipate.
- <u>1.4. Tocopherols</u>, for example α -tocopherol, β -tocopherol, γ -tocopherol, δ -tocopherol and mixtures thereof (Vitamin E).
- 1.5. Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis-(3,6-di-sec-amylphenol), 4,4'-bis-(2,6-dimethyl-4-hydroxyphenyl) disulfide.
- 1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-(α-methylcyclohexyl)phenol], 2,2'-methylenebis(4-methyl-6-cyclohexyl-phenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-(α-methylbenzyl)-4-nonylphenol], 2,2'-methylenebis[6-(α,α-dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methyl-phenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methyl-phenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-

tert-butyl-4-methylphenyl]terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxy-phenyl)butane, 2,2-bis-(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis-(5-tert-butyl-4-hydroxy2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra-(5-tert-butyl-4-hydroxy2-methylphenyl)pentane.

- 1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5di-tert-butyl-4-hydroxybenzylmercaptoacetate.
- 1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis-(3,5-di-tert-butyl-2-hydroxybenzyl)-malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5--methylbenzyl)-malonate, di-dodecylmercaptoethyl-2,2-bis-(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-tetramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.
- 1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.
- 1.10. Triazine Compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

- 1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.
- 1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.
- 1.13. Esters of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris-(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.
- 1.14. Esters of β-(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with monoor polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.
- 1.15. Esters of β-(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris-(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

- 1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris-(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.
- 1.17. Amides of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazine.

1.18. Ascorbic acid (vitamin C)

1.19. Aminic antioxidants, for example N,N'-di-isopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-pphenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-p-phenylenediamine, Nisopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-pphenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, Ncyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxydiphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert-octyldiphenylamine, 4-n-butylaminophenol, 4-butyrylaminophenol, 4-nonanoylamino-phenol, 4-dodecanoylaminophenol, 4-octadecanoylaminophenol, bis(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2methylphenyl)amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, Bis[4-(1',3'-dimethylbutyl)phenyl]amine, tert-octylated N-phenyl-1-naphthylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mixture of

mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphenylamines, a mixture of mono- und dialkylated tert-butyldiphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- und dialkylated tert-butyl/tert-octylphenothiazines, a mixture of mono- und dialkylated tert-octyl-phenothiazines, N-allylphenothiazin, N,N,N',N'-tetraphenyl-1,4-diamino-but-2-ene, N,N-bis(2,2,6,6-tetramethyl-piperid-4-yl-hexamethylenediamine, bis(2,2,6,6-tetramethylpiperid-4-yl)sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol.

2. UV absorbers and light stabilisers

Nickel compounds, for example nickel complexes of 2,2'-thio-bis-[4-(1,1,3,3-tetra-methylbutyl)phenol], such as the 1:1 or 1:2 complex, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of the monoalkyl esters, e.g. the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid, nickel complexes of ketoximes, e.g. of 2-hydroxy-4-methylphenyl undecylketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.

- 3. Metal deactivators, for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl hydrazine, N,N'-bis(salicyloyl) hydrazine, N,N'-bis(3,5-di-tert-butyl-4-hydroxy-phenylpropionyl) hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)-oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenyl-hydrazide, N,N'-diacetyladipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.
- 4. Phosphites and phosphonites, for example triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-ditert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-

butylphenyl) pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)-pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methyl-phenyl)methylphosphite, bis(2,4-di-tert-butyl-6-methylphosphite.

- 5. Hydroxylamines, for example, N,N-dibenzylhydroxylamine, N,N-diethylhydroxylamine, N,N-dioctylhydroxylamine, N,N-diletradecylhydroxylamine, N,N-dihexadecylhydroxylamine, N,N-dioctadecylhydroxylamine, N-hexadecyl-N-octadecylhydroxylamine, N-heptadecyl-N-octadecylhydroxylamine, N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.
- 6. Nitrones, for example, N-benzyl-alpha-phenyl-nitrone, N-ethyl-alpha-methyl-nitrone, N-octyl-alpha-heptyl-nitrone, N-lauryl-alpha-undecyl-nitrone, N-tetradecyl-alpha-tridecyl-nitrone, N-hexadecyl-alpha-pentadecyl-nitrone, N-octadecyl-alpha-heptadecyl-nitrone, N-ocatadecyl-alpha-pentadecyl-nitrone, N-hexadecyl-alpha-heptadecyl-nitrone, N-octadecyl-alpha-pentadecyl-nitrone, N-heptadecyl-alpha-heptadecyl-nitrone, N-octadecyl-alpha-hexadecyl-nitrone, nitrone derived from N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.
- 7. Thiosynergists, for example, dilauryl thiodipropionate or distearyl thiodipropionate.
- 8. Peroxide scavengers, for example esters of β -thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercaptobenzimidazole, zinc dibutyldithiocarbamate, dioctadecyl disulfide, pentaerythritol tetrakis(β -dodecylmercapto)propionate.
- 9. Basic co-stabilisers, for example, melamine, polyvinylpyrrolidone, dicyandiamide, triallyl cyanurate, urea derivatives, hydrazine derivatives, amines,

polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or tin pyrocatecholate.

- 10. Nucleating agents, for example, inorganic substances such as talcum, metal oxides such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates of, preferably, alkaline earth metals; organic compounds such as monoor polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds such as ionic copolymers ("ionomers").
- 11. Fillers and reinforcing agents, for example, calcium carbonate, silicates, glass fibres, glass bulbs, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibers of other natural products, synthetic fibers.
- 12. Other additives, for example, plasticisers, lubricants, emulsifiers, pigments, rheology additives, catalysts, flow-control agents, optical brighteners, flameproofing agents, antistatic agents and blowing agents.
- 13. Benzofuranones and indolinones, for example those disclosed in US-A-4325863, US-A-4338244, US-A-5175312, US-A-5216052, US-A-5252643, DE-A-4316611, DE-A-4316622, DE-A-4316876, EP-A-0589839 or EP-A-0591102 or 3--[4-(2-acetoxyethoxy)phenyl]-5,7-di-tert-butyl-benzofuran-2-one, 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]benzofuran-2-one, 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)benzofuran-2-one], 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one, 3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-tert-butyl-benzofuran-2-one, 3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-tert-butyl-benzofuran-2-one.

The weight ratio of the total amount of components (I), (II) and optionally (X-1) and/or (X-2), as well as optionally component (XX) to the total amount of the conventional additives can be, for example, 100:1 to 1:100.

The examples below illustrate the invention in greater detail. All percentages and parts are by weight, unless stated otherwise.

Stabilizers used in the following Examples I to IV:

(For the polymeric compounds, the mean degree of polymerization is indicated in each case.)

Compound (B-1-a-1):

(CHIMASSORB 944 (RTM))

with b₁ being 4.5.

Compound (B-2-a-1):

(CHIMASSORB 119 (RTM))

Compound (B-7-a-1):

(TINUVIN 622 (RTM))

with n₁ being 5.1.

Compound (X-2-a):

TINUVIN 327 (RTM)

$$CI$$
 N
 OH
 $C(CH_3)_3$
 $C(CH_3)_3$

EXAMPLE I: Light stabilization of polypropylene tapes.

100 parts of polypropylene powder (melt flow index: 3.2 g/10 min at 230°C and 2160 g) are blended in a barrel mixer with 0.05 parts of pentaerythrityl tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate], 0.05 parts of tris[2,4-di-tert-butylphenyl] phosphite and the stabilizer system indicated in Table 1. Then, the blend is compounded in an extruder at temperatures of 180°- 220°C. The granules obtained on extrusion and granulation are transformed into films at 220°-260°C in a second extruder equipped with a flat sheet die. The films are cut into ribbons which are drawn to achieve a stretch ratio of 1:6. The tapes obtained with this procedure are finally 50 µm thick and 2.5 mm wide.

The tapes are mounted without tension on sample holders and exposed to natural weathering in Florida (45° South, direct, approximately 145 kLy/year). Periodically, the tensile strength of the exposed tapes is measured. The received energy (in kLy)

corresponding to a loss of 50 % (E_{50}) of the initial tensile strength or the % retained tensile strength after 320 kLy is a measure for the stabilizing efficiency of the stabilizer system.

The values obtained are summarized in Table 1.

Table 1:

Stabilizer System	E ₅₀ (kLy) to 50% retained tensile strength or % retained tensile strength after 320 kLy
0.1 % of Mg stearate	58
0.05 % of (B-1-a-1) +0.05 % of (B-7-a-1) + 0.1 % of Mg stearate	78 %

EXAMPLE II: Light stabilization of polypropylene homopolymer films.

100 parts of unstabilized polypropylene powder (melt flow index: 3.8 g/10 min at 230°C and 2160 g) are homogenized at 200°C for 10 min in a Brabender plastograph with 0.05 parts of pentaerythrityl-tetrakis{3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate}, 0.10 parts of tris{2,4-di-tert-butylphenyl} phosphite and the stabilizer system indicated in Tables 2a, 2b, 2c and 2d. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 min at 260°C to a 0.5 mm thick film which is cooled immediately to room temperature in a water-cooled press. Samples of 60 mm x 25 mm are cut out of these 0.5 mm films and are exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer. The exposure time corresponding to formation of a carbonyl absorbance of 0.1 in hours $(T_{0.1})$ is a measure for the efficiency of the stabilizer system. The values obtained are summarized in Tables 2a, 2b, 2c and 2d.

Table 2a:

Stabilizer system	To 1 in hours to 0.1 carbonyl absorbance	
0.1 % of Mg stearate + 0.1 % of (X-2-a)	360	
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	1960	

0.1 % of Mg stearate + 0.1 % of (X-2-a)	
0.025 % of (B-2-a-1) + 0.025 % of (B-7-a-1) +	1820
0.1 % of Mg stearate + 0.1 % of (X-2-a)	

Table 2b:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.1 % of Zn stearate + 0.5 % of TiO ₂ (rutile)	430
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	1540
0.1 % of Zn stearate + 0.5 % of TiO ₂ (rutile)	
0.025 % of (B-2-a-1) + 0.025 % of (B-7-a-1) +	1840
0.1 % of Zn stearate + 0.5 % of TiO₂ (rutile)	

Table 2c:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	2020
0.1 % of hydrotalcite (DHT-4A (RTM)) + 0.5 % of TiO ₂ (rutile)	

Table 2d:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	2320
0.1 % of hydrotalcite (DHT-4A (RTM)) + 0.1 % of (X-2-a)	

EXAMPLE III: Light stabilization of polypropylene copolymer films.

100 parts of unstabilized copolypropylene powder (melt flow index: 6 g/10 min at 230°C and 2160 g; 10 % ethylene monomer) are homogenized at 200°C for 10 min in a Brabender plastograph with 0.05 parts of pentaerythrityl-tetrakis{3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate}, 0.10 parts of tris{2,4-di-tert-butylphenyl} phosphite and the stabilizer system indicated in Table 3. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 min at 260°C to a 0.5 mm thick film which is cooled immediately to room temperature in a water-cooled press. Samples of 60 mm x 25 mm are cut out of these 0.5 mm films and are exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer. The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the efficiency of the stabilizer system. The values obtained are summarized in Table 3.

Table 3:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.1 % of hydrotalcite (DHT-4A (RTM)) + 0.5 % of TiO ₂ (rutile)	390
0.05 % of (B-1-a-1) + 0.05 % of (B-7-a-1) +	4000
0.1 % of hydrotalcite (DHT-4A (RTM)) + 0.5 % of TiO₂ (rutile)	
0.05 % of (B-2-a-1) + 0.05 % of (B-7-a-1) +	4960
0.1 % of hydrotalcite (DHT-4A (RTM)) + 0.5 % of TiO ₂ (rutile)	

EXAMPLE IV: Light stabilization of polyethylene HD films.

100 parts of unstabilized high density polyethylene powder (density: 0.964 g cm⁻³, melt flow index: 5.0 g/10 min at 190°C and 2160 g) are homogenized at 180°C for 10 min in a Brabender plastograph with 0.03 parts of octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionate and the stabilizer system indicated in Tables 4a, 4b, 4c, 4d, 4e, 4f, 4g, 4h and 4i. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 min at 210°C to a 0.5 mm thick film which is cooled immediately to room temperature in a water-cooled press. Samples of 60 mm x 25 mm are cut out of these 0.5 mm films and are exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer. The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the efficiency of the stabilizer mixture. The values obtained are summarized in Table 4a, 4b, 4c, 4d, 4e, 4f, 4g, 4h and 4i.

Table 4a:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	18060
0.1 % of Mg stearate	

Table 4b:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	18055
0.1 % of hydrotalcite (DHT-4A (RTM))	

Table 4c:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	22210
0.05 % of Mg stearate + 0.05 % of Ca stearate	

Table 4d:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	15680
0.05 % of hydrotalcite (DHT-4A (RTM)) + 0.1 % of Ca stearate	

Table 4e:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	23040 .
0.05 % of hydrotalcite (DHT-4A (RTM)) + 0.1 % of Mg stearate	

Table 4f:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	>28700
0.1 % of Mg stearate + 0.5 % of TiO ₂ (rutile)	

Table 4g:

Stabilizer system	T _{0 1} in hours
	to 0.1 carbonyl

	absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	>28700
0.05 % of Mg stearate + 0.5 % of TiO ₂ (rutile) + 0.05 % of Ca stearate	

Table 4h:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	>28700
0.05 % of hydrotalcite (DHT-4A (RTM)) + 0.5 % of TiO ₂ (rutile) +	
0.05 % of Ca stearate	

Table 4i:

Stabilizer system	T _{0.1} in hours to 0.1 carbonyl absorbance
0.025 % of (B-1-a-1) + 0.025 % of (B-7-a-1) +	>28700
0.05 % of hydrotalcite (DHT-4A (RTM)) + 0.5 % of TiO₂ (rutile) +	
0.05 % of Mg stearate	

Claims:

- 1. A stabilizer mixture containing
- (I) two different sterically hindered amine compounds, and
- (II) at least one compound selected from the group consisting of an organic salt of Zn, an inorganic salt of Zn, Zn oxide, Zn hydroxide, an organic salt of Mg, an inorganic salt of Mg, Mg oxide and Mg hydroxide.
- 2. A stabilizer mixture according to claim 1 wherein the two different sterically hindered amine compounds of component (I) are selected from the group consisting of

 $(\alpha-1)$ a compound of the formula (A-1)

$$\begin{bmatrix} H_3C & CH_3 & O \\ E_1 & N & O \end{bmatrix} = \begin{bmatrix} H_3C & CH_3 & O \\ H_3C & CH_3 & D \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & D \\ H_3C & CH_3 & D \end{bmatrix}$$

$$\begin{bmatrix} H_3C & CH_3 & D \\ H_3C & CH_3 & D \end{bmatrix}$$

in which

 E_1 is hydrogen, C_1 - C_8 alkyl, O', -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl,

m₁ is 1, 2 or 4,

if m_1 is 1, E_2 is C_1 - C_{25} alkyl,

if m₁ is 2, E₂ is C₁-C₁₄alkylene or a group of the formula (a-l)

$$- \begin{matrix} \downarrow \\ C \\ E_3 \end{matrix} \qquad \begin{matrix} E_5 \\ E_6 \end{matrix}$$
 OH (a-I)

wherein E_3 is C_1 - C_{10} alkyl or C_2 - C_{10} alkenyl, E_4 is C_1 - C_{10} alkylene, and E_5 and E_6 independently of one another are C_1 - C_4 alkyl, cyclohexyl or methylcyclohexyl, and

if m₁ is 4, E₂ is C₄-C₁₀alkanetetrayl;

 $(\alpha-2)$ a compound of the formula (A-2)

in which

two of the radicals E7 are -COO-(C1-C20alkyl), and two of the radicals E7 are a group of the formula (a-II)

$$- COO - H_3C CH_3$$

with E₈ having one of the meanings of E₁;

 $(\alpha-3)$ a compound of the formula (A-3)

in which

E₉ and E₁₀ together form C₂-C₁₄alkylene, E₁₁ is hydrogen or a group -Z₁-COO-Z₂, Z₁ is C₂-C₁₄alkylene, and

Z₂ is C₁-C₂₄alkyl, and

E₁₂ has one of the meanings of E₁;

 $(\alpha-4)$ a compound of the formula (A-4)

wherein

the radicals E_{13} independently of one another have one of the meanings of E_1 , the radicals E_{14} independently of one another are hydrogen or C_1 - C_{12} alkyl, and E_{15} is C_1 - C_{10} alkylene or C_3 - C_{10} alkylidene;

$(\alpha-5)$ a compound of the formula (A-5)

wherein

the radicals E_{16} independently of one another have one of the meanings of E_1 ;

$(\alpha-6)$ a compound of the formula (A-6)

$$E_{17} \longrightarrow O \qquad H_3C \qquad CH_3$$

$$N \longrightarrow E_{18} \qquad (A-6)$$

$$O \qquad H_3C \qquad CH_3$$

in which

E₁₇ is C₁-C₂₄alkyl, and

E₁₈ has one of the meanings of E₁;

$(\alpha-7)$ a compound of the formula (A-7)

$$E_{19} \longrightarrow E_{21}$$

$$(A-7)$$

in which

 E_{19} , E_{20} and E_{21} independently of one another are a group of the formula (a-III)

wherein E22 has one of the meanings of E1;

 $(\alpha-8)$ a compound of the formula (A-8)

wherein

the radicals E_{23} independently of one another have one of the meanings of E_1 , and E_{24} is hydrogen, C_1 - C_{12} alkyl or C_1 - C_{12} alkoxy;

 $(\alpha-9)$ a compound of the formula (A-9)

$$\begin{bmatrix} H_3C & CH_3 & O \\ E_{\overline{25}} & N & N & E_{26} \\ H_3C & CH_3 & m_2 \end{bmatrix}$$
 (A-9)

wherein

m₂ is 1, 2 or 3,

E25 has one of the meanings of E1, and

when
$$m_2$$
 is 1, E_{26} is a group $---$ CH₂CH₂-NH- $\left\langle \right\rangle$,

when m_2 is 2, E_{26} is C_2 - C_{22} alkylene, and when m_2 is 3, E_{26} is a group of the formula (a-IV)

wherein the radicals E_{27} independently of one another are C_2 - C_{12} alkylene, and the radicals E_{28} independently of one another are C_1 - C_{12} alkyl or C_5 - C_{12} cycloalkyl;

 $(\alpha-10)$ a compound of the formula (A-10)

wherein

the radicals E_{29} independently of one another have one of the meanings of E_1 , and E_{30} is C_2 - C_{22} alkylene, C_5 - C_7 cycloalkylene, C_1 - C_4 alkylenedi(C_5 - C_7 cycloalkylene), phenylene or phenylenedi(C_1 - C_4 alkylene);

 $(\beta-1)$ a compound of the formula (B-1)

in which

 R_1 , R_3 , R_4 and R_5 independently of one another are hydrogen, C_1 - C_{12} alkyl, C_5 - C_{12} cycloalkyl, C_1 - C_4 -alkyl-substituted C_5 - C_{12} cycloalkyl, phenyl, phenyl which is substituted by -OH and/or C_1 - C_{10} alkyl; C_7 - C_9 phenylalkyl, C_7 - C_9 phenylalkyl which is substituted on the phenyl radical by -OH and/or C_1 - C_{10} alkyl; or a group of the formula (b-l)

 R_2 is C_2 - C_{18} alkylene, C_5 - C_7 cycloalkylene or C_1 - C_4 alkylenedi(B_5 - C_7 cycloalkylene), or

the radicals R_1 , R_2 and R_3 , together with the nitrogen atoms to which they are bonded, perform a 5- to 10-membered heterocyclic ring, or

 R_4 and R_5 , together with the nitrogen atom to which they are bonded, form a 5- to 10-membered heterocyclic ring,

 R_6 is hydrogen, C_1 - C_8 alkyl, O° , -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl, and

 b_1 is a number from 2 to 50,

with the proviso that at least one of the radicals R_1 , R_3 , R_4 and R_5 is a group of the formula (b-I);

(β-2) a compound of the formula (B-2)

wherein

 R_7 and R_{11} independently of one another are hydrogen or C_1 - C_{12} alkyl, R_8 , R_9 and R_{10} independently of one another are C_2 - C_{10} alkylene, and X_1 , X_2 , X_3 , X_4 , X_5 , X_6 , X_7 and X_8 independently of one another are a group of the formula (b-II),

$$\begin{array}{c|c} & H_3C & CH_3 \\ \hline & N & R_{13} \\ \hline & R_{12} & H_3C & CH_3 \end{array} \tag{b-II)}$$

in which R_{12} is hydrogen, C_1 - C_{12} alkyl, C_5 - C_{12} cycloalkyl, C_1 - C_4 alkyl-substituted C_5 - C_{12} cycloalkyl, phenyl, -OH- and/or C_1 - C_{10} alkyl-substituted phenyl, C_7 - C_9 phenylalkyl which is substituted on the phenyl radical by -OH and/or C_1 - C_{10} alkyl; or a group of the formula (b-l) as defined above, and R_{13} has one of the meanings of R_6 ;

$(\beta-3)$ a compound of the formula (B-3)

in which

 R_{14} is C_1 - C_{10} alkyl, C_5 - C_{12} cycloalkyl, C_1 - C_4 alkyl-substituted C_5 - C_{12} cycloalkyl, phenyl or C_1 - C_{10} alkyl-substituted phenyl,

R₁₅ is C₃-C₁₀alkylene,

 R_{16} has one of the meanings of $R_{6},$ and

b₂ is a number from 2 to 50;

$(\beta-4)$ a compound of the formula (B-4)

in which

 R_{17} and R_{21} independently of one another are a direct bond or a -N(X₉)-CO-X₁₀-CO-N(X₁₁)-group, where X₉ and X₁₁ independently of one another are hydrogen, C₁-C₈alkyl,

C₅-C₁₂cycloalkyl, phenyl, C₇-C₉phenylalkyl or a group of the formula (b-l),

X₁₀ is a direct bond or C₁-C₄alkylene,

R₁₈ has one of the meanings of R₆,

R₁₉, R₂₀, R₂₃ and R₂₄ independently of one another are hydrogen, C₁-C₃₀alkyl,

C5-C12cycloalkyl or phenyl,

 R_{22} is hydrogen, C_1 - C_{30} alkyl, C_5 - C_{12} cycloalkyl, phenyl, C_7 - C_9 phenylalkyl or a group of the formula (b-l), and

b₃ is a number from 1 to 50;

 $(\beta-5)$ a compound of the formula (B-5)

(B-5)

in which

 R_{25} , R_{26} , R_{27} , R_{28} and R_{29} independently of one another are a direct bond or C_1 - C_{10} alkylene,

 R_{30} has one of the meanings of R_6 , and b_4 is a number from 1 to 50;

 $(\beta-6)$ a product (B-6) obtainable by reacting a product, obtained by reaction of a polyamine of the formula (B-6-1) with cyanuric chloride, with a compound of the formula (B-6-2)

$$H_2N - (CH_2)_{\overline{b''_5}} NH - (CH_2)_{\overline{b'''_5}} NH - (CH_2)_{\overline{b'''_5}} NH_2$$
 (B-6-1)

$$H \longrightarrow R_{31}$$
 (B-6-2)
$$H_3C \longrightarrow R_{32}$$
 CH_3 CH_3

in which

b'₅, b"₅ and b"'₅ independently of one another are a number from 2 to 12, R_{31} is hydrogen, C_1 - C_{12} alkyl, C_5 - C_{12} cycloalkyl, phenyl or C_7 - C_9 phenylalkyl, and R_{32} has one of the meanings of R_6 ;

 $(\beta-7)$ a compound of the formula (B-7)

wherein A_1 is hydrogen or C_1 - C_4 alkyl, A_2 is a direct bond or C_1 - C_{10} alkylene, and n_1 is a number from 2 to 50;

 $(\beta-8)$ at least one compound of the formulae (B-8-a) and (B-8-b)

$$\begin{array}{c|c}
CH & CH_2 \\
CH_2 & CH_2
\end{array}$$

$$\begin{array}{c|c}
CH_2 & CH_3 \\
CH_3 & CH_3
\end{array}$$

$$\begin{array}{c|c}
CH_3 & CH_3 \\
CH_3 & CH_3
\end{array}$$

$$\begin{array}{c|c}
CH_3 & CH_3
\end{array}$$

wherein n₂ and n₂* are a number from 2 to 50;

(β-9) a compound of the formula (B-9)

(B-9)

wherein A_3 and A_4 independently of one another are hydrogen or C_1 - C_8 alkyl, or A_3 and A_4 together form a C_2 - C_{14} alkylene group, and the variables n_3 independently of one another are a number from 1 to 50; and

(β-10) a compound of the formula (B-10)

wherein n₄ is a number from 2 to 50,

A₅ is hydrogen or C₁-C₄alkyl,

the radicals A_6 and A_7 independently of one another are C_1 - C_4 alkyl or a group of the formula (b-l),

with the proviso that at least 50 % of the radicals A_7 are a group of the formula (b-1).

- 3. A stabilizer mixture according to claim 2, wherein the two different sterically hindered amine compounds of component (I) are selected from the group consisting of the classes $(\alpha-1)$, $(\alpha-2)$, $(\alpha-3)$, $(\alpha-4)$, $(\alpha-5)$, $(\alpha-6)$, $(\alpha-7)$, $(\alpha-8)$, $(\alpha-9)$ and $(\alpha-10)$.
- 4. A stabilizer mixture according to claim 2, wherein

the two different sterically hindered amine compounds of component (I) are selected from the group consisting of the classes (β -1), (β -2), (β -3), (β -4), (β -5), (β -6), (β -7), (β -8), (β -9) and (β -10).

- 5. A stabilizer mixture according to claim 2, wherein one of the two different sterically hindered amine compounds of component (I) is selected from the group consisting of the classes (α -1), (α -2), (α -3), (α -4), (α -5), (α -6), (α -7), (α -8), (α -9) and (α -10), and the other of the two different sterically hindered amine compounds of component (I) is selected from the group consisting of the classes (β -1), (β -2), (β -3), (β -4), (β -5), (β -6), (β -7), (β -8), (β -9) and (β -10).
- 6. A stabilizer mixture according to claim 2, wherein one of the two different sterically hindered amine compounds of component (I) is selected from the class (β -1).
- 7. A stabilizer mixture according to claim 2, wherein one of the two different sterically hindered amine compounds of component (I) is selected from the class (β -1), and the other of the two different sterically hindered amine compounds of component (I) is selected from the class (α -1) or (β -7).
- 8. A stabilizer mixture according to claim 2, wherein one of the two different sterically hindered amine compounds of component (I) is selected from the class (β -7), and the other of the two different sterically hindered amine compounds of component (I) is selected from the class (β -2).
- 9. A stabilizer mixture according to claim 2, wherein the two different sterically hindered amine compounds of component (I) are selected from different classes.
- 10. A stabilizer mixture according to claim 2, wherein

m₁ is 1, 2 or 4,

if m₁ is 1, E₂ is C₁₂-C₂₀alkyl,

if m₁ is 2, E₂ is C₂-C₁₀alkylene or a group of the formula (a-l)

E₃ is C₁-C₄alkyl,

E4 is C1-C6alkylene, and

E₅ and E₆ independently of one another are C₁-C₄alkyl, and

if m₁ is 4, E₂ is C₄-C₈alkanetetrayl;

two of the radicals E7 are -COO-(C10-C15alkyl), and

two of the radicals E7 are a group of the formula (a-II);

E₉ and E₁₀ together form C₉-C₁₃alkylene,

E₁₁ is hydrogen or a group -Z₁-COO-Z₂,

Z₁ is C₂-C₆alkylene, and

Z₂ is C₁₀-C₁₆alkyl;

E₁₄ is hydrogen, and

E₁₅ is C₂-C₆alkylene or C₃-C₅alkylidene;

E₁₇ is C₁₀-C₁₄alkyl;

E₂₄ is C₁-C₄alkoxy;

m₂ is 1, 2 or 3,

when m_2 is 1, E_{26} is a group $--CH_2CH_2-NH-$

when m₂ is 2, E₂₆ is C₂-C₆alkylene, and

when m₂ is 3, E₂₆ is a group of the formula (a-IV)

the radicals E27 independently of one another are C2-C6alkylene, and

the radicals E28 independently of one another are C1-C4alkyl or C5-C8cycloalkyl; and

E₃₀ is C₂-C₈alkylene;

R₁ and R₃ independently of one another are a group of the formula (b-I),

R₂ is C₂-C₈alkylene,

R₄ and R₅ independently of one another are hydrogen, C₁-C₁₂alkyl, C₅-C₈cycloalkyl or a group of the formula (b-l), or the radicals R₄ and R₅, together with the nitrogen atom to which they are bonded, form a 5- to 10-membered heterocyclic ring, and

b₁ is a number from 2 to 25;

R₇ and R₁₁ independently of one another are hydrogen or C₁-C₄alkyl,

R₈, R₉ and R₁₀ independently of one another are C₂-C₄alkylene, and

X₁, X₂, X₃, X₄, X₅, X₆, X₇ and X₈ independently of one another are a group of the formula

(b-II),

R₁₂ is hydrogen, C₁-C₄alkyl, C₅-C₈cycloalkyl or a group of the formula (b-I);

R₁₄ is C₁-C₄alkyl,

R₁₅ is C₃-C₆alkylene, and

b₂ is a number from 2 to 25;

R₁₇ and R₂₁ independently of one another are a direct bond or a group

-N(X₉)-CO-X₁₀-CO-N(X₁₁)-,

X₉ and X₁₁ independently of one another are hydrogen or C₁-C₄alkyl,

X₁₀ is a direct bond,

R₁₉ and R₂₃ are C₁-C₂₅alkyl or phenyl,

R₂₀ and R₂₄ are hydrogen or C₁-C₄alkyl,

R₂₂ is C₁-C₂₅alkyl or a group of the formula (b-l), and

b₃ is a number from 1 to 25;

R₂₅, R₂₆, R₂₇, R₂₈ and R₂₉ independently of one another are a direct bond or

C₁-C₄aikylene, and

b₄ is a number from 1 to 25;

b'5, b"5 and b"5 independently of one another are a number from 2 to 4, and

R₃₁ is hydrogen, C₁-C₄alkyl, C₅-C₈cycloalkyl, phenyl or benzyl;

A₁ is hydrogen or methyl,

A₂ is a direct bond or C₂-C₆alkylene, and

n₁ is a number from 2 to 25;

n₂ and n₂* are a number from 2 to 25;

A₃ and A₄ independently of one another are hydrogen or C₁-C₄alkyl, or A₃ and A₄ together form a C₉-C₁₃alkylene group, and

the variables n₃ independently of one another are a number from 1 to 25;

n₄ is a number from 2 to 25,

A₅ and A₆ independently of one another are C₁-C₄alkyl, and

A₇ is C₁-C₄alkyl or a group of the formula (b-l)

with the proviso that at least 50 % of the radicals A₇ are a group of the formula (b-I).

11. A stabilizer mixture according to claim 1, wherein

the two different sterically hindered amine compounds of component (I) are selected from the group consisting of the compounds of the formulae (A-1-a), (A-1-b), (A-1-c), (A-1-d),

(A-2-a), (A-3-a), (A-3-b), (A-4-a), (A-4-b), (A-5), (A-6-a), (A-7), (A-8-a), (A-9-a), (A-9-b), (A-9-c), (A-10-a), (B-1-a), (B-1-b), (B-1-c), (B-1-d), (B-2-a), (B-3-a), (B-4-a), (B-4-b) and (B-4-c), a product (B-6-a) and the compounds of the formulae (B-7-a), (B-8-a), (B-8-b), (B-9-a) and (B-10-a);

$$\begin{array}{c|c} & CH_3 \\ \hline E_1 & N \\ \hline \end{array} \begin{array}{c} O \\ \hline C \\ \hline \end{array} \begin{array}{c} O \\ \hline C \\ \hline \end{array} \begin{array}{c} O \\ \hline C \\ \hline \end{array} \begin{array}{c} O \\ \end{array} \end{array} \begin{array}{c} O \\ \end{array} \end{array} \begin{array}{c} O \\ \end{array} \end{array} \begin{array}{c} O \\ \end{array} \begin{array}{c} O \\ \end{array} \begin{array}{c} O \\ \end{array} \end{array} \begin{array}{c} O \\ \end{array} \begin{array}{c} O \\ \end{array} \end{array} \begin{array}{c} O \\ \end{array} \begin{array}{c} O \\ \end{array} \begin{array}{c} O \\ \end{array} \end{array} \begin{array}{c} O \\$$

$$H_3C$$
 CH_3
 CH_3

$$\begin{bmatrix} H_3C & CH_3 & O \\ E_1 - N & O - C \end{bmatrix} \xrightarrow{C_4H_9} C C(CH_3)_3$$

$$C_4 = CH_2 \xrightarrow{C_4H_9} C C(CH_3)_3$$

$$C_4 = CH_2 \xrightarrow{C_4H_9} C C(CH_3)_3$$

$$C_4 = CH_3 \xrightarrow{C_4H_9} C C(CH_3)_3$$

wherein E_1 is hydrogen, C_1 - C_8 alkyl, O', -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl;

in which two of the radicals E_7 are -COO- $C_{13}H_{27}$ and

of E₁;

$$(CH2)9 O C = O$$

$$H3C O CH3$$

$$H3C O CH3$$

$$CH3 CH3$$

$$\begin{array}{c} CH_{2} \\ CH_{2}O \\ CH_{2} \\ CH_{3}C \\ H_{3}C \\ CH_{3} \\ CH_{3} \\ CH_{3} \\ CH_{3} \end{array} \qquad \begin{array}{c} CH_{2}CH_{2} \\ CH_{2}CH_{2} \\ CH_{3} \\ CH_{4} \\ CH_{5} \\ CH$$

wherein E₁₂ has one of the meanings of E₁;

wherein E₁₃ has one of the meanings of E₁;

wherein E_{16} has one of the meanings of $E_{1;}$

wherein E_{18} has one of the meanings of $E_{1;}$

$$E_{19} \longrightarrow E_{21}$$

$$O \longrightarrow E_{21}$$

in which E_{19} , E_{20} and E_{21} independently of one another are a group of the formula (a-III)

wherein E_{22} has one of the meanings of $E_{1;}$

wherein E_{23} has one of the meanings of $E_{1;}$

$$\begin{array}{c|c} H_3C & CH_3 & O \\ \hline E_{\overline{25}} & N & N - CH_2CH_{\overline{2}} - NH - \\ \hline \\ H_3C & CH_3 \end{array}$$
 (A-9-a)

wherein E_{25} has one of the meanings of $E_{1;}$

wherein E_{29} has one of the meanings of $E_{1;}$

wherein b₁ is a number from 2 to 20 and R₆ is hydrogen, C₁-C₈alkyl, O', -OH, -CH₂CN, C₁-C₁₈alkoxy, C₅-C₁₂cycloalkoxy, C₃-C₆alkenyl, C₇-C₉phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C₁-C₄alkyl; or C₁-C₈acyl;

wherein R_{13} has one of the meanings of R_{6} ,

wherein b_2 is a number from 2 to 20 and R_{16} has one of the meanings of R_6 ;

wherein b₃ is a number from 1 to 20 and R₁₈ has one of the meanings of R₆;

wherein b4 is a number from 1 to 20 and R30 has one of the meanings of R6;

a product (B-6-a) obtainable by reacting a product, obtained by reaction of a polyamine of the formula (B-6-1-a) with cyanuric chloride, with a compound of the formula (B-6-2-a)

$$H_2N - (CH_2) - NH - (CH_2) - NH - (CH_2) - NH_2$$
 (B-6-1-a)

$$H \longrightarrow C_4H_9$$
 (B-6-2-a)
$$H_3C \longrightarrow CH_3$$

$$H_3C \longrightarrow CH_3$$

$$H_{32}$$

in which R_{32} has one of the meanings of R_6 .

wherein n_1 is a number from 2 to 20;

$$\begin{array}{c|c}
\hline
CH & CH_2 & O \\
\hline
CH_2 & O & O \\
\hline
CH_2 & O & O \\
\hline
CH_3 & CH_3 & CH_3 \\
\hline
H_3C & N & CH_3 \\
\hline$$

wherein n₂ and n₂* are a number from 2 to 20;

(B-9-a)

wherein the variables n₃ independently of one another are a number from 1 to 20;

wherein n_4 is a number from 2 to 20, and at least 50 % of the radicals A_7 are a group of the formula (b-I)

wherein R_6 is hydrogen, C_1 - C_8 alkyl, O° , -OH, -CH₂CN, C_1 - C_{18} alkoxy, C_5 - C_{12} cycloalkoxy, C_3 - C_6 alkenyl, C_7 - C_9 phenylalkyl unsubstituted or substituted on the phenyl by 1, 2 or 3 C_1 - C_4 alkyl; or C_1 - C_8 acyl,

and the remaining radicals A7 are ethyl.

12. A stabilizer mixture according to claim 11 wherein

the two different sterically hindered amine compounds of component (I) are

- a compound of the formula (A-1-b) wherein E₁ is hydrogen, and a compound of the formula (B-1-a) wherein R₆ is hydrogen;
- 2) a compound of the formula (B-1-a) wherein R₆ is hydrogen, and a compound of the formula (B-7-a); or

- 3) a compound of the formula (B-2-a) wherein R₁₃ is methyl, and a compound of the formula (B-7-a).
- 13. A stabilizer mixture according to claim 2 wherein E_1 , E_8 , E_{12} , E_{13} , E_{16} , E_{18} , E_{22} , E_{23} , E_{25} , E_{29} , R_6 , R_{13} , R_{16} , R_{18} , R_{30} and R_{32} are hydrogen, C_1 - C_4 alkyl, C_1 - C_{10} alkoxy, cyclohexyloxy, allyl, benzyl or acetyl.
- 14. A stabilizer mixture according to claim 11 wherein E_1 , E_8 , E_{12} , E_{13} , E_{16} , E_{18} , E_{22} , E_{23} , E_{25} , E_{29} , R_6 , R_{13} , R_{16} , R_{18} , R_{30} and R_{32} are hydrogen or methyl and E_1 and R_6 additionally are C_1 - C_8 alkoxy.
- 15. A stabilizer mixture according to claim 1, wherein the compound of component (II) is selected from the group consisting of Mg carboxylates, Zn carboxylates, Mg oxides, Zn oxides, Mg hydroxides, Zn hydroxides, Mg carbonates and Zn carbonates.
- 16. A stabilizer mixture according to claim 1, which additionally contains as a further component
- (X-1) a pigment or
- (X-2) an UV absorber or
- (X-3) a pigment and an UV absorber.
- 17. A stabilizer mixture according to claim 1, which additionally contains as a further component
- (XX) an organic salt of Ca, an inorganic salt of Ca, Ca oxide or Ca hydroxide.
- 18. A composition comprising an organic material subject to degradation induced by light, heat or oxidation and a stabilizer mixture according to claim 1.
- 19. A composition according to claim 18 wherein the organic material is a synthetic polymer.
- 20. A composition according to claim 18 wherein the organic material is a polyolefin.

- 21. A composition according to claim 18 wherein the organic material is polyethylene, polypropylene, a polyethylene copolymer or a polypropylene copolymer.
- 22. A method for stabilizing an organic material against degradation induced by light, heat or oxidation, which comprises incorporating into the organic material a stabilizer mixture according to claim 1.

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Abstract

A stabilizer mixture containing

- (1) two different sterically hindered amine compounds, and
- (II) at least one compound selected from the group consisting of an organic salt of Zn, an inorganic salt of Zn, Zn oxide, Zn hydroxide, an organic salt of Mg, an inorganic salt of Mg, Mg oxide and Mg hydroxide.

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